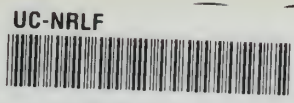


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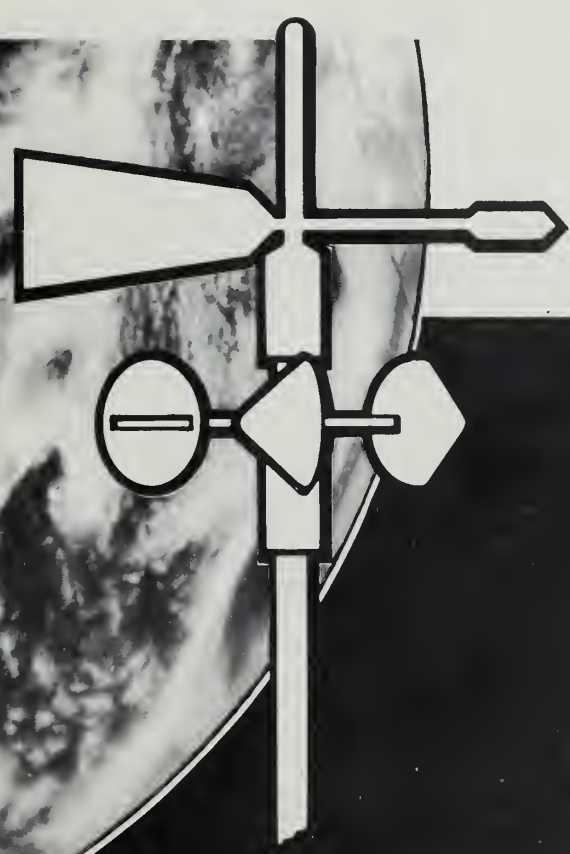
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MEASUREMENTS OF ULTRAVIOLET RADIATION IN A POLLUTED ATMOSPHERE

by
Kinsell L. Coulson

Scientific Report No. 1
Grant No. 5 R01 AP00742-02



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IN A POLLUTED ATMOSPHERE

by

Kinsell L. Coulson
Professor of Meteorology
Department of Agricultural Engineering

Grant No. 5 R01 AP00742-02
U. S. Public Health Service

December, 1969

Abstract

Results from measurements of the relative intensity and degree of polarization of ultraviolet light over the hemisphere of the sunlit sky for the case of a polluted atmosphere in Los Angeles are shown and compared to similar data for a clear natural atmosphere and to results computed for a pure Rayleigh atmospheric model. It is shown that pollution brings about an increase of intensity over most of the sky by a factor of two or more, a pronounced decrease of intensity near the horizons, and a significant change of the intensity pattern over the sky hemisphere. The degree of polarization is decreased by a factor of more than two by pollution, but the polarization pattern is not greatly changed otherwise. The positions of the neutral points of skylight polarization in a polluted atmosphere are shifted from their measured positions in a clear atmosphere, the Babinet point being shifted by as much as 10° toward the sun and the Arago point by up to 8° toward the anti-sun by the effects of pollution.



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I. Introduction

A. Background and objective.

The objective of the present investigation is to determine the effects of atmospheric pollution on the ultraviolet radiation regime of the atmosphere. The general method of approach is to make measurements of the intensity, polarization, and flux of ultraviolet radiation in a heavily polluted atmosphere and compare those results both with similar measurements taken in a very clear atmosphere and with the results obtained from radiative transfer theory.

The atmosphere of Los Angeles was chosen for the polluted case, and the heavy smog season of September-October was chosen as the period for measurements. Consequently, field trips to Los Angeles for measuring the ultraviolet radiation in smog were made during the period September 20 to October 26, 1968, and from September 22 to October 3, 1969. We are indebted to Mr. John Mills, Director of Technical Services, Los Angeles Air Pollution Control District for permission to use the roof of the Air Pollution Control Building in downtown Los Angeles as a site for the measurements, an adjacent instruments room for installation of the recording equipment, and numerous other privileges and facilities which added immeasurably to both the success and satisfaction in the project.

The 1968 time period was a particularly fortunate choice, as four days of heavy smog were encountered (September 26, 27, October 25, 26), and on one day (September 23) there was very little smog in the Basin. Thus, measurements for the two extreme conditions were obtained. Conditions during the 1969 trip were less desirable for the measurements. The stratus overcast was very persistent in the area during the first

half of the period, and although stratus was minimal during the last half, the smog was confined to a relatively thin layer near the ground and the intensity was extremely variable. Thus, the Los Angeles measurements during the 1969 period are somewhat less definitive than those obtained during 1968.

Comparison measurements were made on the University of California campus at Davis during periods carefully selected for the clarity of the atmosphere. On two days on which measurements were made (November 12 and December 17, 1968), the visibility was estimated as eighty miles, and on one day (April 10, 1969) the visibility was twenty to thirty miles. The sky was free of clouds on all three days.

Data resulting from the application of radiative transfer theory to the case of a pure molecular (Rayleigh) atmosphere were taken from Coulson et al (1960), and those for a turbid atmosphere were obtained from various sources, including Coulson (1969), Kano (1964, 1968), Deirmendjian (1964), and others.

B. Instrumentation and mode of operation.

1. Polarizing multi-channel radiometer.

The polarizing multi-channel radiometer is an instrument with somewhat unique features which was designed by the Principal Investigator, and fabricated in the shops at the University of California, Davis. Basically, it measures the relative intensity, degree of plane polarization, and orientation of the plane of polarization of solar radiation in any one of six different spectral intervals, two of the intervals being in the ultraviolet, three in the visible, and one in the near infrared

regions of the spectrum. The relative response to solar radiation of the system with the six interference filters in combination with an S-20 response of the photomultiplier tube is shown in Figure 1, and filter specifications are listed in Table I.

TABLE I. Specifications of the Optical Interference Filters Used in the Polarizing Multi-channel Radiometer

Filter Number	Wavelength of Peak Transmission	Transmission at the Maximum	Bandwidth at Half Transmission Point
1	0.320 μ	21.3%	0.0290 μ
2	0.365	25.0	0.0410
3	0.451	38.1	0.0200
4	0.552	38.0	0.0180
5	0.652	25.1	0.0170
6	0.795	31.0	0.0200

The optical system, shown schematically in Figure 2, consists of a collimating tube which admits radiation from a 1.5° half-angle cone, light chopper, rotating analyzer (Glan-Thompson polarizing prism), set of spectral filters, two sets of neutral density filters, diverging lens, and end-on photomultiplier tube (EMI 9558B). With this optical system, the output signal from the photomultiplier is a composite A.C. voltage consisting in general of a low frequency sine wave modulation (introduced by rotation of the analyzer) and a high frequency square wave signal (resulting from action of the light chopper), of varying magnitude. On rectification, a D.C. signal, consisting of a mean D.C. voltage on which is superimposed the low frequency sine wave modulation, is obtained. The mean

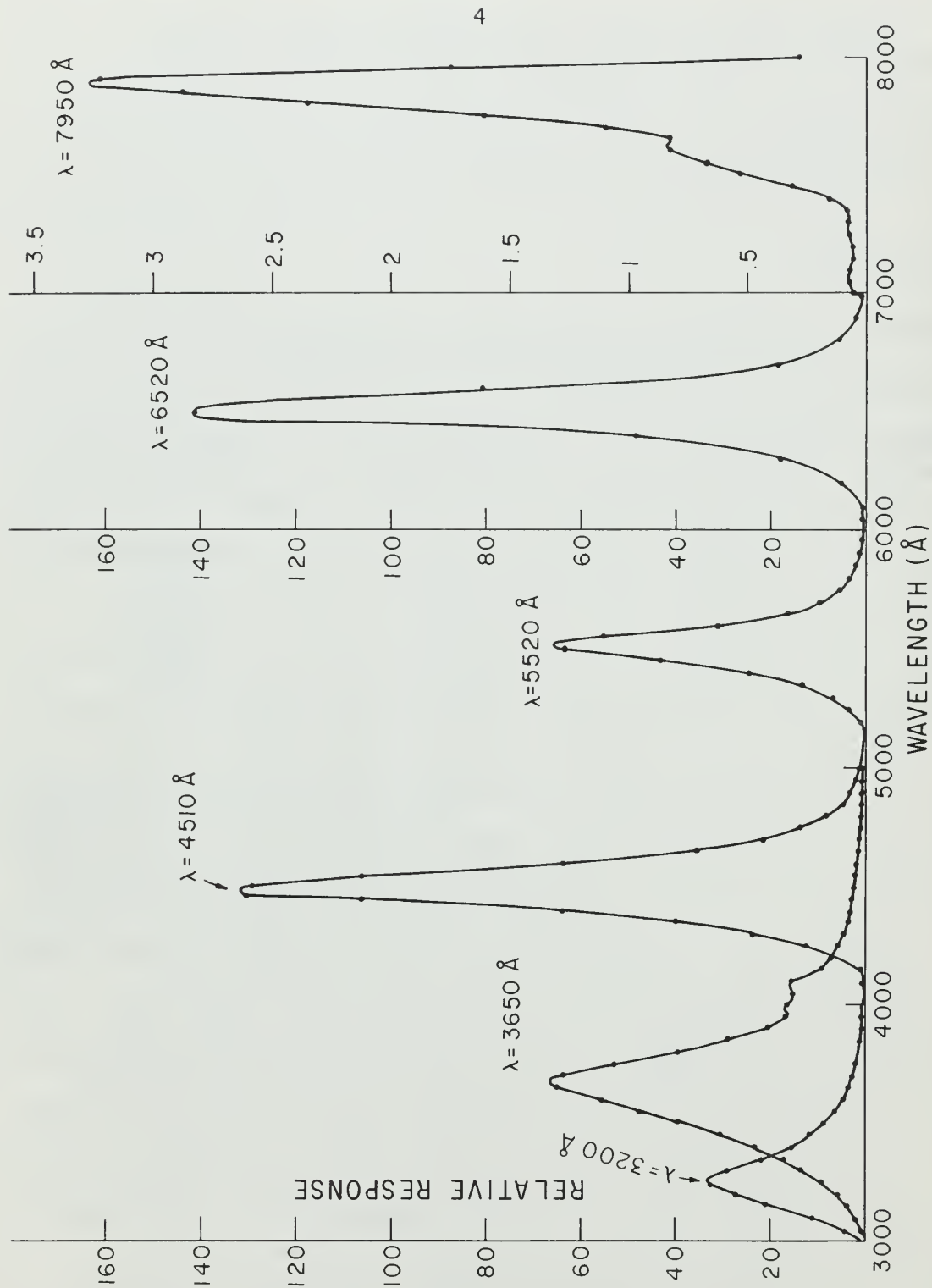


Fig. 1 Relative response to solar radiation of the system containing six optical interference filters in combination with the S-20 photocathode of the photomultiplier tube.

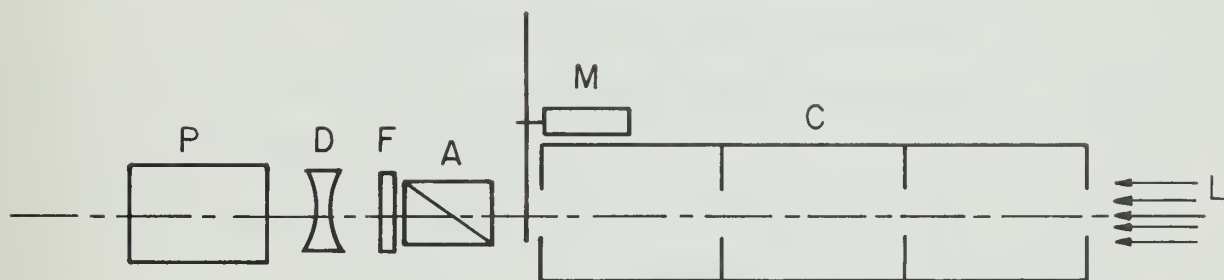


Fig. 2 Schematic diagram of the optical system of the polarizing multi-channel radiometer.

value of the signal is proportional to the total intensity of light incident on the photomultiplier tube, the relative amplitude of the superimposed sine wave is proportional to the degree of plane polarization, and the phase of the sine wave is determined directly by the orientation of the plane of polarization. Thus the radiation field, except for any elliptic polarization, can be determined completely from the output signal of the instrument. Although the instrument could be made to measure elliptic polarization by the insertion of a retardation plate in the optical system, this adds complications and is not considered necessary, as both scattering theory and previous measurements have shown the elliptic polarization to be extremely small in the atmosphere.

The general relation describing the intensity $I(\delta, \psi)$ incident on the photomultiplier tube, as a function of retardation δ of the retardation plate and angle ψ of the plane of transmission of the analyzer, is given by:

$$I(\delta, \psi) = I_e \cos^2 \psi + I_r \sin^2 \psi + 1/2(U \cos \delta - V \sin \delta) \sin 2\psi \quad (1)$$

where I_e and I_r are the intensity components along the vertical and horizontal directions, respectively, U is the Stokes parameter describing the angle χ between the plane of polarization and the vertical direction by the relation

$$\tan 2\chi = \frac{U}{I_e - I_r} \quad (2)$$

and V is the Stokes parameter specifying the ellipticity of the incident radiation. Since we do not include a retardation plate in the system, we set $\delta = 0$, in which case Eq. (1) reduces to

$$I(\psi) = I_e \cos^2 \psi + I_r \sin^2 \psi + 1/2 U \sin 2\psi \quad (3)$$

Thus we have three unknowns (I_e , I_r , U), which can theoretically be determined by three measurements of I at properly chosen values of ψ , from which the total intensity of I , degree of plane polarization P , and orientation of the plane of polarization χ , can be determined by Eq. (2) and the relations

$$I = I_e + I_r \quad (4)$$

$$P = \frac{[(I_e - I_r)^2 + U^2]^{1/2}}{I_e + I_r} \quad (5)$$

It is well known, however, that the accuracy of the measurements can be increased by increasing the number of independent measurements. In fact, if n is the number of independent measurements, all of comparable accuracy, then the standard deviation of the various parameters is proportional to $(n)^{-1/2}$. In order to take advantage of this characteristic, the number of measurements made in one rotation of the analyzer ($0 \leq \psi \leq 2\pi$) was increased to twelve, which were spaced at intervals of ψ of 30° . Then the twelve values of intensity $I(\psi_i)$ producing the corresponding signals S_i , $i = 1, 12$ can be resolved into Fourier components given by the series (Conrad and Pollack, 1950):

$$I(\psi) = p_0 + p_1 \cos \psi + p_2 \cos 2\psi + \dots + q_1 \sin \psi + q_2 \sin 2\psi + \dots \quad (6)$$

where the mean is:

$$p_0 = \frac{1}{12} \sum_{i=1}^{12} S_i \quad (7)$$

and the coefficients for the k -th harmonic ($k \leq 5$) are:

$$\left. \begin{aligned} p_k &= \frac{1}{6} \sum_{i=1}^{12} S_i \cos (i k \alpha) \\ q_k &= \frac{1}{6} \sum_{i=1}^{12} S_i \sin (i k \alpha) \end{aligned} \right\} \quad (8)$$

The Stokes parameters are given in terms of these coefficients by:

$$\left. \begin{aligned} I &= 2 p_0 \\ Q &= 2 p_2 \\ U &= 2 q_2 \end{aligned} \right\} \quad (9)$$

from whence the degree and plane of polarization are:

$$\left. \begin{aligned} P &= \frac{(p_2^2 + q_2^2)^{1/2}}{p_0} \\ \chi &= 1/2 \tan^{-1} (q_2/p_2) \end{aligned} \right\} \quad (10)$$

The physical reason that all of the polarization information is contained in the second harmonic is that for given conditions the same amount of light is transmitted by the prism oriented at $\psi + \pi$ as at orientation ψ .

Two methods of recording the signal from the polarizing radio-meter are used simultaneously during operations. A standard recording potentiometer on which the signal is recorded continuously is used to monitor the instrument operation and for making notes of filter changes, orientation of the instrument, and other relevant information. The twelve voltages S_i resulting from one rotation of the analyzer are measured by a digital voltmeter and impressed on a digital incremental magnetic tape system. Processing of the data, including the introduction of calibration coefficients,

filter factors, and corrections for intensity changes during a measurement series, as well as the performance of the harmonic analysis, is done by electronic computer from the magnetic tape. The strip chart is used only for checking the results.

The normal mode of operation of the instrument is to sweep through vertical planes, from 10° above one horizon through the zenith to 10° above the opposite horizon, with measurements at 10° intervals during the sweep. In order to correct for any dark current voltages from the system and for changes of overall intensity during the nine minutes required for one sweep, additional measurements of the dark current voltages and the light intensity at the zenith were made before and after each sweep. Additional measurements were also made with the instrument oriented at 3° above and below the sun and at 5° above the horizon at each end of the sweep. The largest number of sweeps were made in the vertical plane through the azimuth of the sun, as this is the plane containing the neutral points (points of zero polarization) and the maximum of the polarization field, so the information density is greater in this plane than in other planes of the hemisphere. However, to obtain intensity and polarization data for plotting hemispheric maps of these quantities, it was necessary to make measurements at the additional azimuths of 45° , 90° , and 135° from the azimuth of the sun. Symmetry around the principal plane (vertical plane through the sun) was assumed.

During the 1969 field trip to Los Angeles some auxiliary instrumentation was used for measuring the total flux of ultraviolet radiation. Two small portable ultraviolet flux meters ("Blak-Ray UV Meter" - Ultraviolet Products, Inc., San Gabriel, California) were used for determining the absolute energy flux in the ultraviolet region. One of the two (Model J-221) was kindly loaned to the project by Mr. Norman White of the National Air Pollution Control Administration, Cincinnati, Ohio, and the other (Model J-225) was purchased on this project. Unfortunately both of these instruments were saturated by the radiant flux during the period from mid-morning to mid-afternoon, but frequent measurements were made with them during the morning or afternoon hours of the measurement period.

In addition, two ultraviolet flux meters especially constructed for this project were used for continuous recording of the relative flux of radiation in the wavelength interval shown by the filter transmission curve of Figure 3. One of the two is based on the ordinary type of integrating sphere, in which radiation is admitted through an aperture in the sphere wall and reflected from the highly reflecting inside walls of the sphere, thereby producing a quasi-isotropic radiation field inside the sphere. This radiation field, the intensity of which is proportional to the entering flux, is viewed by the photometer from a direction at a right angle to that of the entrance aperture. Thus the signal from the photometer is proportional to the normal (vertical) component of the energy flux across a plane horizontal surface of unit area. This normal flux F_n is composed of two parts, one due to the approximately parallel

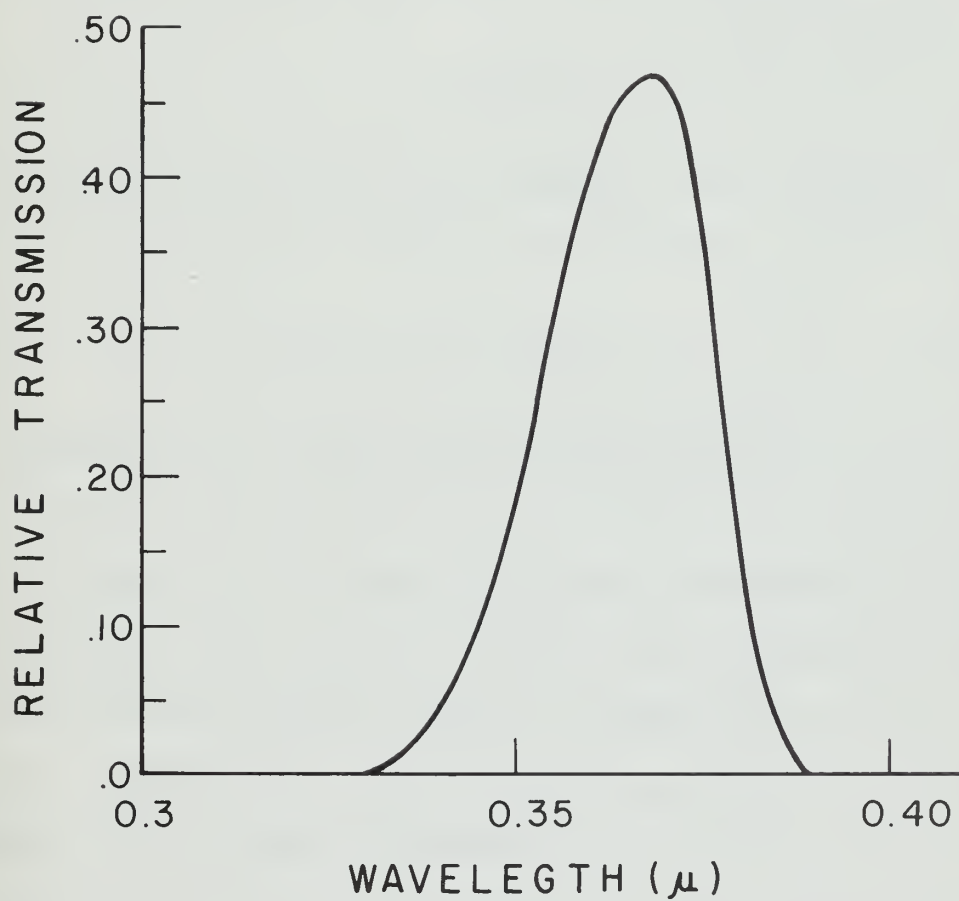


Fig. 3 Transmission curve of the filters used in the two integrating sphere ultraviolet flux meters.

solar radiation F_{nD} which is transmitted directly through the atmosphere, and the other due to the diffuse radiation F_{nd} of the skylight. The fluxes are additive, so we have the normal flux from the upward hemisphere as:

$$F_n = F_{nD} + F_{nd} \quad (11)$$

By the Bouguer-Lambert law, the directly transmitted part of the extra-atmosphere flux F_0 is:

$$F_{nD}(\tau; \theta_0) = F_0 \cos \theta_0 e^{-\tau \sec \theta_0} \quad (12)$$

where τ is normal optical thickness of the atmosphere and θ_0 is zenith angle of the sun. The normal flux of diffuse radiation is:

$$F_{nd} = \int_0^{2\pi} \int_0^{\pi/2} I(\theta, \phi) \sin \theta \cos \theta d\theta d\phi \quad (13)$$

where $I(\theta, \phi)$ is the intensity of diffuse radiation at zenith angle θ and azimuth ϕ .

The other instrument developed on this project is designed to measure the ultraviolet flux through a spherical volume, regardless of its direction of propagation. Although complete directional independence has not yet been achieved in the actual instrument, it is well to look at the relations governing its operation. By an argument analogous to that leading to Equations (11), (12), and (13), it is easy to see that the flux F_s from the upward hemisphere through a spherical volume of unit cross sectional area is:

$$F_s = F_{sD} + F_{sd} \quad (14)$$

where

$$F_{sD} = F_0 e^{-\tau \sec \theta_0} \quad (15)$$

and

$$F_{SD} = \int_0^{2\pi} \int_0^{\pi/2} I(\theta, \phi) \sin \theta \, d\theta \, d\phi \quad (16)$$

Certain characteristics can be seen from Equations (12), (13), (15), and (16). First, for the sun at the zenith, $\cos \theta_0 = 1$ and $F_{nD} = F_{SD}$. Secondly, as θ_0 approaches $\pi/2$ the fluxes F_{nD} and F_{SD} both approach zero, but F_{nD} approaches zero faster than does F_{SD} . Finally, Equations (13) and (16) show that for the case of the diffuse skylight being isotropically distributed over the hemisphere of the sky,

$$F_{nD} = 1/2 F_{SD} \quad (17)$$

So far we have not considered the radiation which is reflected from the ground. The surface-reflected radiation constitutes a flux directed upward through both the spherical volume and horizontal surface, and is also responsible for an enhancement of the downward diffuse intensity of the skylight $I(\theta, \phi)$. If we denote the surface reflected radiation by subscript g , then we have

$$F_{ng} = \int_0^{2\pi} \int_0^{\pi/2} I_g(\theta, \phi) \sin \theta \cos \theta \, d\theta \, d\phi \quad (18)$$

and

$$F_{sg} = \int_0^{2\pi} \int_0^{\pi/2} I_g(\theta, \phi) \sin \theta \, d\theta \, d\phi \quad (19)$$

For practical purposes we can assume the ground surface to be approximated reasonably closely by an ideal Lambert surface of total reflectance (albedo) A . For this case I_g is isotropically distributed

over the hemisphere and is given by the relation:

$$\pi I_g = A (F_{nD} + F_{nd}) \quad (20)$$

The fluxes from the downward hemisphere are then

$$F_{ng} = A (F_{nD} + F_{nd}) \quad (21)$$

and

$$F_{sg} = 2A (F_{nD} + F_{nd}) \quad (22)$$

Backscattering of this surface-reflected radiation by the overlying atmosphere adds a small component I_d^1 to the intensity of diffuse skylight. For the case of a pure molecular atmosphere and Lambert surface reflection, we can express the normal component of the additional small flux as (Deirmendjian and Sekera - 1954):

$$F_{nd}^1 = \frac{\pi F_o \mu_o A \bar{s}}{2 (1 - A \bar{s})} [\gamma_e (\mu_o) + \gamma_r (\mu_o)] \quad (23)$$

Here $\mu_o = \cos \theta_o$, $\bar{s}$ is a known function of optical thickness τ , and $\gamma_e (\mu_o)$ and $\gamma_r (\mu_o)$ are functions for which tabulations are available. Finally, if the backscattered radiation is isotropically distributed over the sky we have

$$F_{sd}^1 = 2 F_{nd}^1 \quad (24)$$

A summation of the various components yields the following total fluxes, F_n being directed downward through a horizontal surface of unit area and F_s including radiation which is proceeding in all directions through the spherical volume of unit cross sectional area:

$$F_n = F_{nD} + F_{nd} + F_{nd}^1 \quad (25)$$

$$F_s = F_{sD} + F_{sd} + 2 A (F_{nD} + F_{nd}) + 2 F_{nd}^1 \quad (26)$$

II. Measurements of Skylight Intensity

The distribution of the intensity of diffusely scattered solar radiation over the dome of the sky has been measured by several authors in the

past. Volz and Bullrich (1961) made measurements of skylight intensity and polarization in relatively broad spectral regions from the near ultraviolet ($\lambda = 0.37\mu$) to the near infrared ($\lambda = 1.76\mu$) in Mainz, Germany, and Bullrich et al (1966a, 1966b, 1968) have made spectral measurements of skylight in the clear atmospheres of Greenland, Hawaii, and New Mexico. DeBary (1964) made measurements at three wavelength ranges in the visible region for comparison with computations for various turbidity conditions. A general discussion of the problem has been given by Feigelson et al (1960).

Skylight intensity measurements in cases of heavily polluted atmospheres are sparse. The atmospheric aerosol which restricted visibility to 1.6 km during the measurements by deBary (1964) at the University of California, Los Angeles, was apparently fog instead of typical smog. The absence of smog in downtown Los Angeles at the time would indicate such to be the case. (UCLA is about 10 miles toward the coast from downtown Los Angeles). Holzworth and Rao (1965) obtained a series of measurements, also in Los Angeles, of the polarization of skylight in three spectral intervals in the visible for various atmospheric conditions and locations in the basin. Their analysis was concentrated mainly on the behavior of the neutral points and the characteristics of the maximum of the polarization field. Unfortunately, their report does not include data on the intensity of skylight.

The skylight intensity and degree of polarization has been measured so far in this continuing study on fourteen days in downtown Los Angeles, (September 22, 23, 24, 25, 26, 27; October 25, 26, 1968, and September 25, 26, 27, 28, 30; October 1, 1969), and on three particularly clear days (November 12, December 17, 1968; and April 10, 1969) on the campus of

the University of California, Davis. The smog conditions of Los Angeles were variable during the periods of measurement, as can be seen by the estimates of horizontal visibility, kindly made available by personnel of the Los Angeles Air Pollution Control District and the California Air Pollution Control Board, shown as Figure 4. The visibility estimates are made on an hourly schedule during normal working hours from the same building on which our measurements were made. We concentrated on measurements during the period from daylight until solar noon, in order to take advantage of the most intense smog conditions. The measurements at Davis were made on exceptionally clear days. Estimates of horizontal visibility for the three days are the following: November 12, 1968 - 80 miles; December 17, 1968 - 80 miles; April 10, 1969 - 30 miles.

For purposes of this report we have chosen to concentrate mostly on measurements on clear and smoggy days in Los Angeles. This assures that the underlying surface effects are uniform in the data, whereas the surface reflection properties are quite different in the vicinity of Davis. The intensity distribution as measured during five sweeps in the principal plane (vertical plane through the sun) at $\lambda = 3650\text{\AA}$ on the clear day (September 23, 1968) in Los Angeles is shown in Figure 5. The distributions at $\lambda = 3650\text{\AA}$ are typical of those at $\lambda = 3200\text{\AA}$ in Los Angeles and at both 3650Å and 3200Å in Davis. In the interest of brevity these other cases are not shown. The general shape of the curves is characteristic of pure molecular (Rayleigh) scattering in many respects. The existence of a broad but well defined minimum at about 90° from the position of the sun, a higher intensity at the azimuth of the sun ($\phi = 0^\circ$)* than at the

* The azimuth designation used here is chosen so as to make a clear physical picture. The indicated azimuth is the angle between the azimuth of the direction viewed by the photometer and the azimuth of the sun.

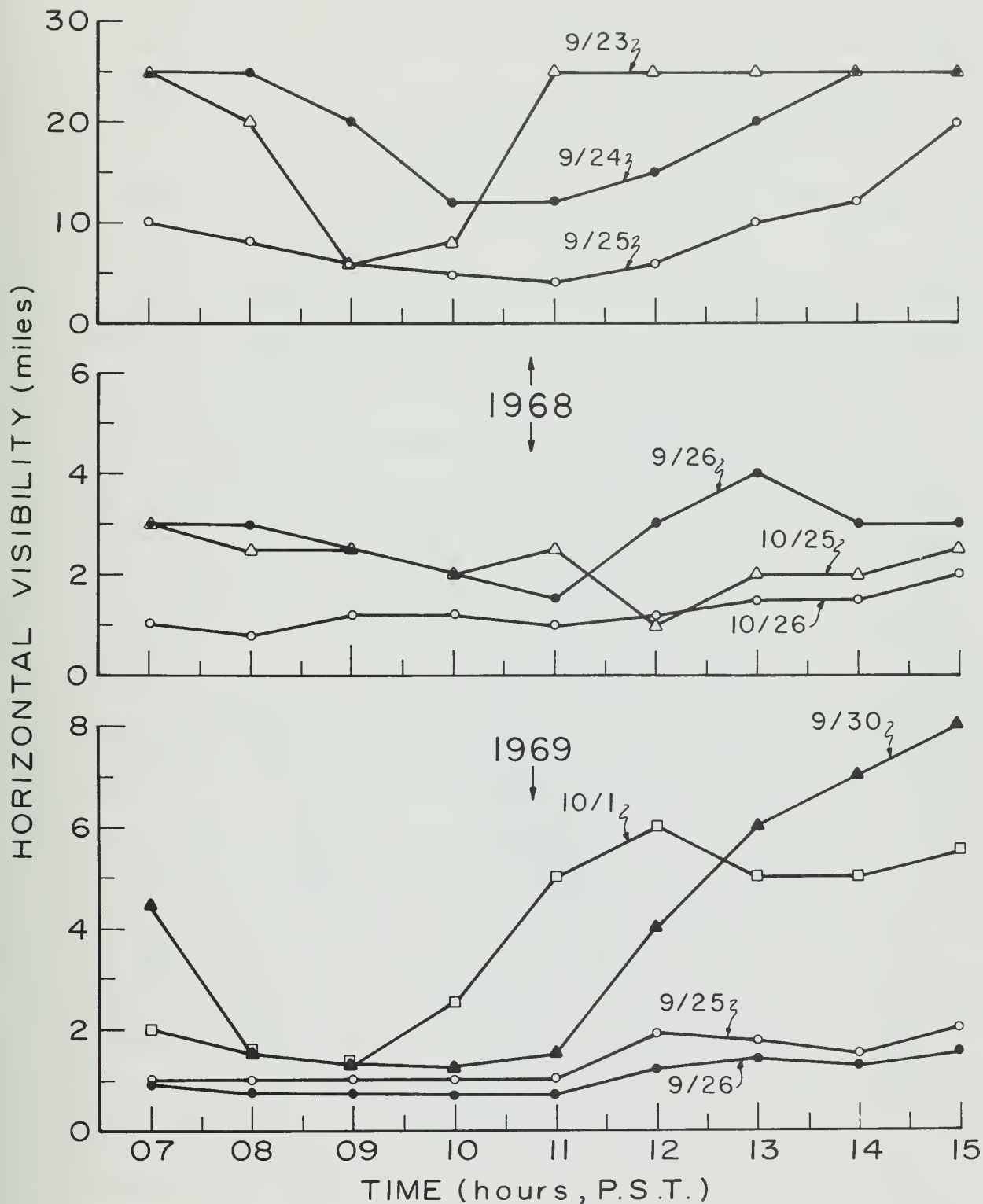


Fig. 4 Horizontal visibility in downtown Los Angeles for the 1968 and 1969 periods of measurements. (data from California Air Pollution Control Board and Los Angeles Air Pollution Control District)

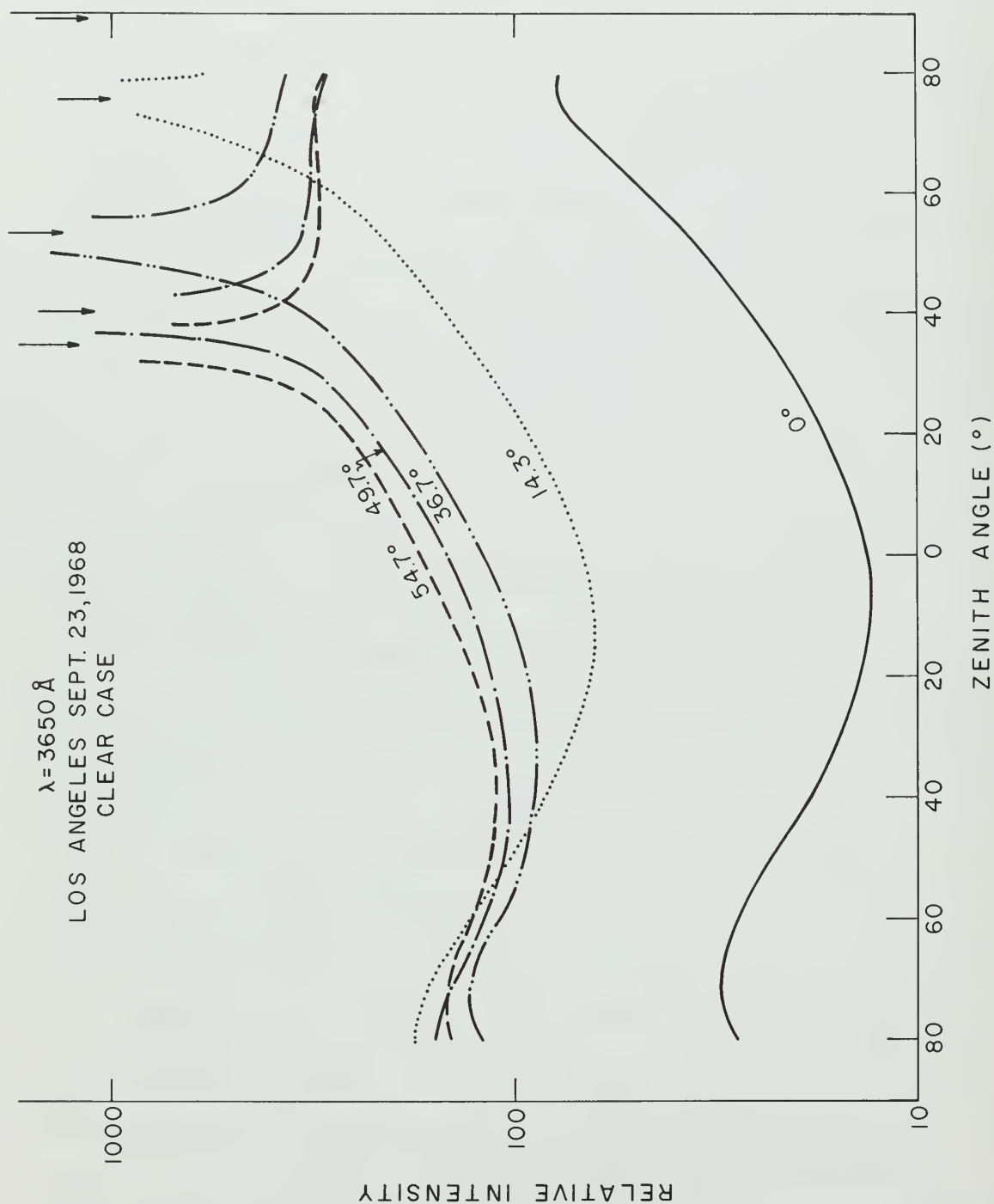


Fig. 5 Relative intensity of skylight as measured in a relatively clear atmosphere in Los Angeles at five different sun elevations (Principal plane, $\lambda = 3650 \text{ \AA}$)

opposite azimuth ($\phi = 180^\circ$), and a slight amount of horizon darkening at short wavelengths are all seen in the intensity distribution for a Rayleigh atmosphere. Similarly, an increase of intensity with increasing sun elevation, the increase being especially pronounced at low sun elevations, and the general shift of the pattern with the sun's position are features seen in perfectly clear atmospheres.

The main feature shown by Figure 5 which does not occur in a Rayleigh atmosphere is the pronounced increase of intensity in the vicinity of the sun. This solar aureole, as it is usually called, is due to the well known strong forward scattering typical of Mie type particles.* Such particles are always present in the natural atmosphere, even under the clearest conditions. The brightness and angular distribution with respect to the solar disk is a function of the number and type of Mie particles in the atmosphere. Although the Los Angeles atmosphere was relatively clear during the period of the measurements shown in Figure 5, the well developed aureole indicates the existence of a considerable turbidity component. The aureole for measurements in Davis (not shown here) is not as well developed as that of Los Angeles, but it is still evident even for the case in which the horizontal visibility was 80 miles.

The equivalent curves of intensity distribution in the principal plane are shown in Figure 6 for the case of heavy smog (September 26, 1968). An idea of the early morning conditions in the Los Angeles area on that date can be obtained from the picture of Figure 7. This picture was taken from the measurement site at 0600 PST, at which time the sun elevation was approximately 1° . The existence of smog is obvious from the picture, and

* Mie particles, named after Gustav Mie who developed the applicable scattering theory, are particles whose diameter is comparable to or larger than the wavelength of the radiation.

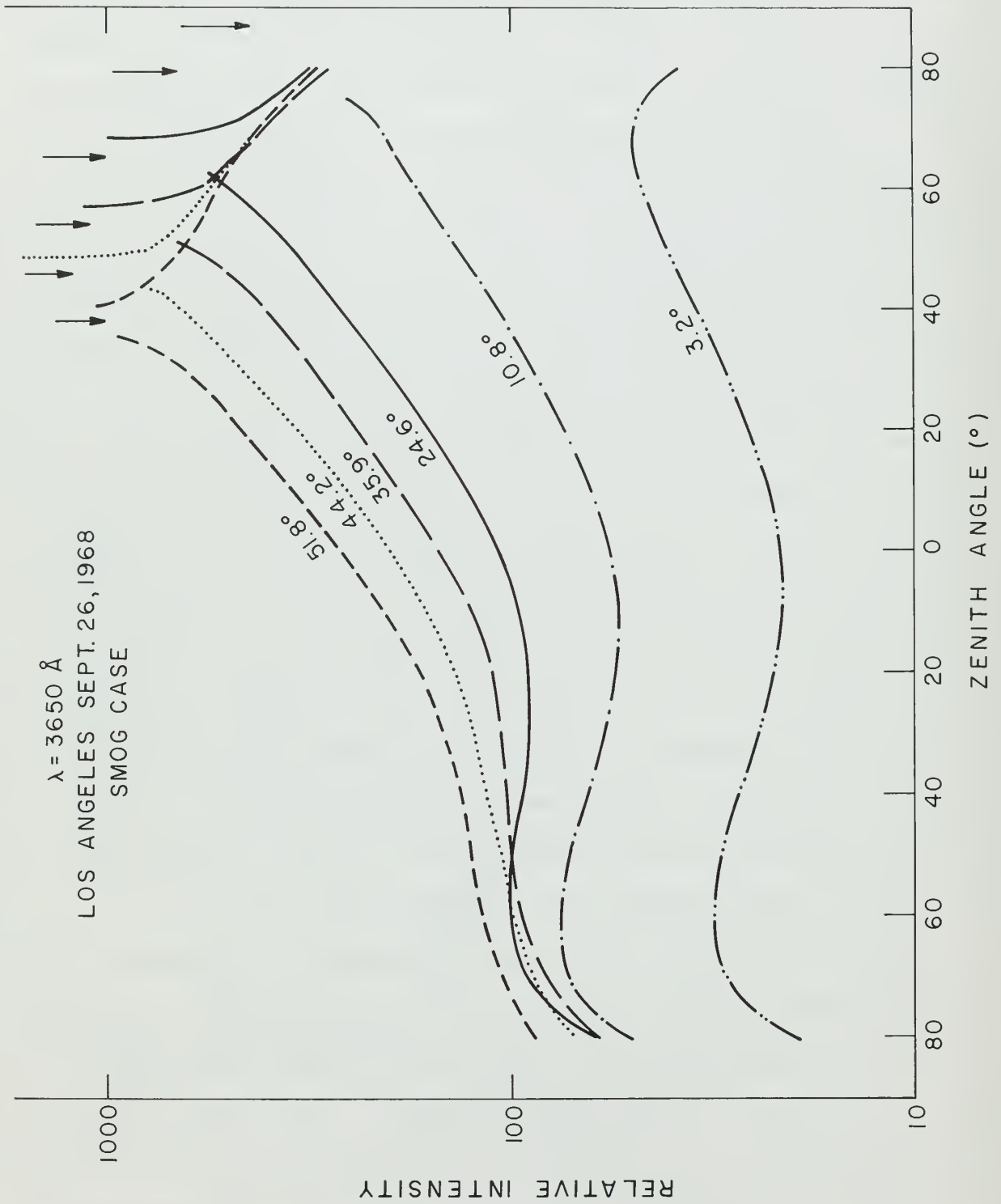


Fig. 6 Relative intensity of skylight as measured in a polluted atmosphere in Los Angeles at six different sun elevations (Principal plane, $\lambda = 3650\text{\AA}$)

Fig. 7 Photograph showing the smog conditions in Los Angeles during the early morning of September 26, 1968 (the day on which many of the data shown in this report were taken.

on the basis of visual observations on this and other days during the period, the smog conditions on September 26 seemed to be typical. However, in the early morning the smog seemed to be confined mainly to the lower levels. The sky overhead was quite blue, and the typical brown color existed only near the horizons. The lower curve of Figure 6, which was taken about ten minutes after the picture of Figure 7, shows a definite minimum near the zenith direction, although the minimum is less pronounced than that for the clear case.

Corroborative evidence that the smog layer was of small vertical extent in the early morning is given by the distribution of polarization, which is discussed in a later section, and by the time lapse movies taken during the period of measurements. It appeared that the smog layer had settled to the low levels during the stable atmospheric conditions and minimal traffic of the hours of darkness, but as the traffic built up one could see smog coming up from between the buildings of the city. The denser smog layer would first obscure the base of a distant building, while leaving the top still plainly visible. As time progressed, surface heating and increased input of pollution combined to thicken the layer, resulting in a gradual obscuration of the tops of buildings, a further enhancement of the brown colored region near the horizons, and eventually a virtual disappearance of the blue color of the sky. This general regime was seen repeatedly during the several days of the measurement period.

The additional curves of Figure 6 show how this progressive buildup of pollution causes a redistribution of the skylight intensity. The solar aureole gradually broadens out from the solar disk, the broadening being due to scattering of the direct solar radiation

by increasing numbers of particles, and more particularly by multiple scattering among the numerous particles themselves. The net result is that by mid-forenoon the solar aureole covers the entire sky, thereby erasing the minimum in the intensity distribution and causing a monotonic increase of intensity all the way from the horizon to the disk of the sun.

In order to show the effect of smog on the intensity distribution over the entire hemisphere* of the sky, we include three series of diagrams, with three diagrams in each series, of the theoretical and measured intensity for various cases. The resulting nine diagrams are Figures 8 through 16. The three diagrams of each series are at comparable sun elevations, the sun elevations for the three series being at about 12° , 36° , and 53° .

Isopleths of intensity for the case of a Rayleigh atmosphere and low sun elevation are shown in Figure 8. (The diagrams for the Rayleigh atmosphere are for $\lambda = 3120\text{\AA}$ whereas the others are for $\lambda = 3650\text{\AA}$. This discrepancy is of no particular consequence, as the Rayleigh patterns are almost identical at the two wavelengths.) The features of the Rayleigh pattern, with which the other cases can be compared, are the existence and positions of the intensity minimum centered at about 10° from the zenith, the two maxima in the vertical plane at 60° to 70° from the zenith, the position and magnitude of the generally bright area above the horizon and the extent of horizon darkening. As can be seen from Figure 8, all of these features are definite and well developed for the case of low sun elevation.

* Only one half of the hemisphere is shown; the other half is a mirror image of this one. Symmetry around the principal plane is assumed.

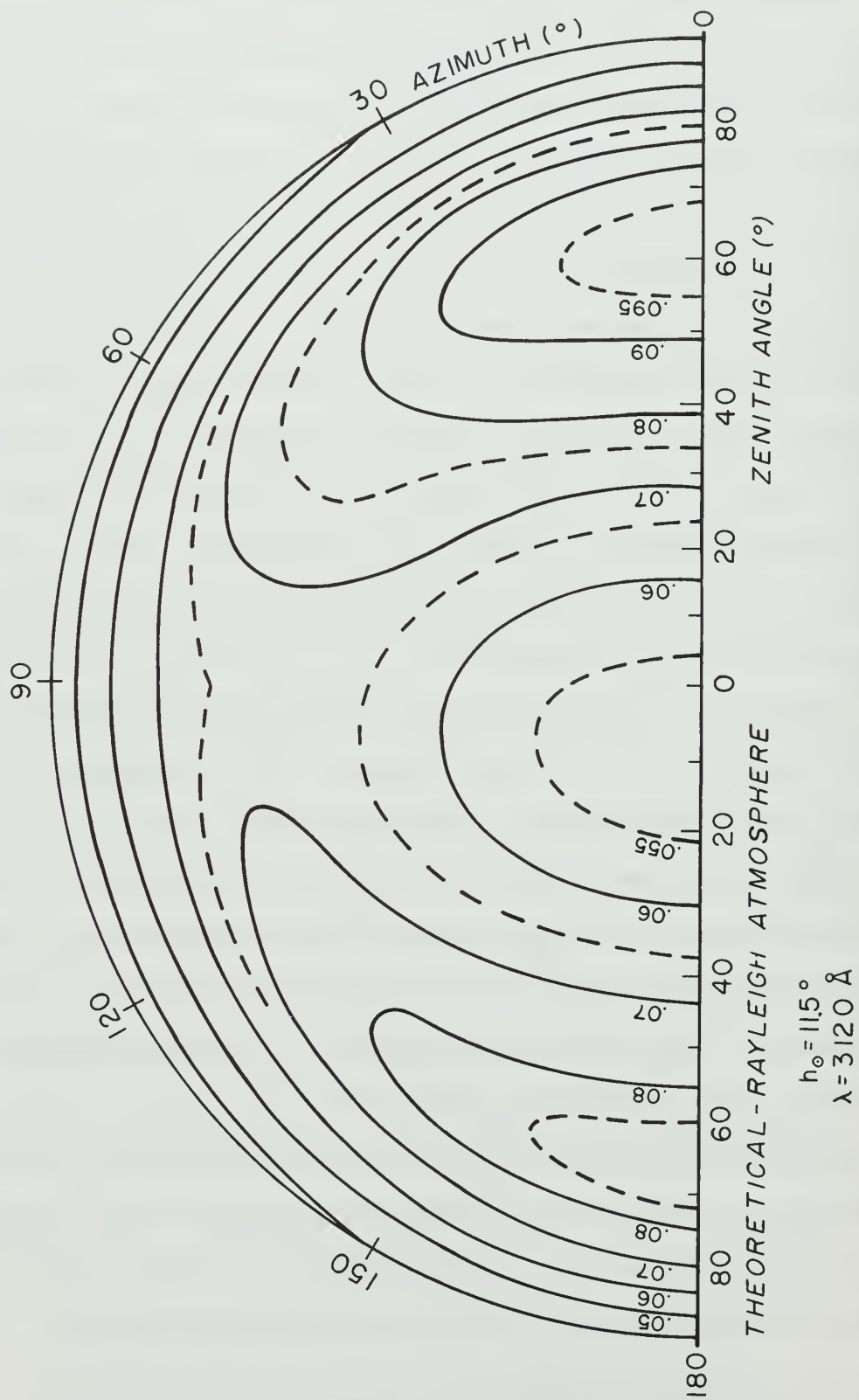


Fig. 8 Intensity distribution over the hemisphere of the sky as computed for a pure molecular atmosphere (Sun elevation 11.5° , $\lambda = 3120\text{\AA}$)

Figure 9 shows the equivalent case for the clear atmosphere of Los Angeles. Although all of the above features are present, they vary in relative importance. Large particle scattering makes the maximum in the region of the sun very pronounced, but that in the $\phi = 180^\circ$ azimuth is weak. Limb darkening is less pronounced than for the Rayleigh case, indicating that there are sufficient Mie particles in the long pathlength near the horizon to add significantly to the brightness. This feature is often observed in the natural atmosphere.

The angular size of the solar aureole is considerably enlarged in the case of smog, as shown in Figure 10. Horizon darkening is much more pronounced at the azimuth opposite the sun ($\phi = 180^\circ$) than in either of the other two cases, a feature which indicates that smog absorbs strongly at these ultraviolet wavelengths. It is obvious from visual observation that such is the case for visible radiation also. While the minima in the $\phi = 180^\circ$ azimuth still exist, they are weak.

The equivalent cases for a sun elevation of about 36° are shown as Figures 11, 12, and 13. The Rayleigh pattern is not changed greatly from the previous case. All of the features mentioned show up very well, the one significant change being a general shift of the pattern toward the horizon opposite the sun. This is to be expected, since the scattering pattern is primarily dependent on scattering angle. A buildup of particulate content during the forenoon is evident, even for the clear atmosphere, as shown by Figure 12. The solar aureole is of greater relative importance than in the previous case.

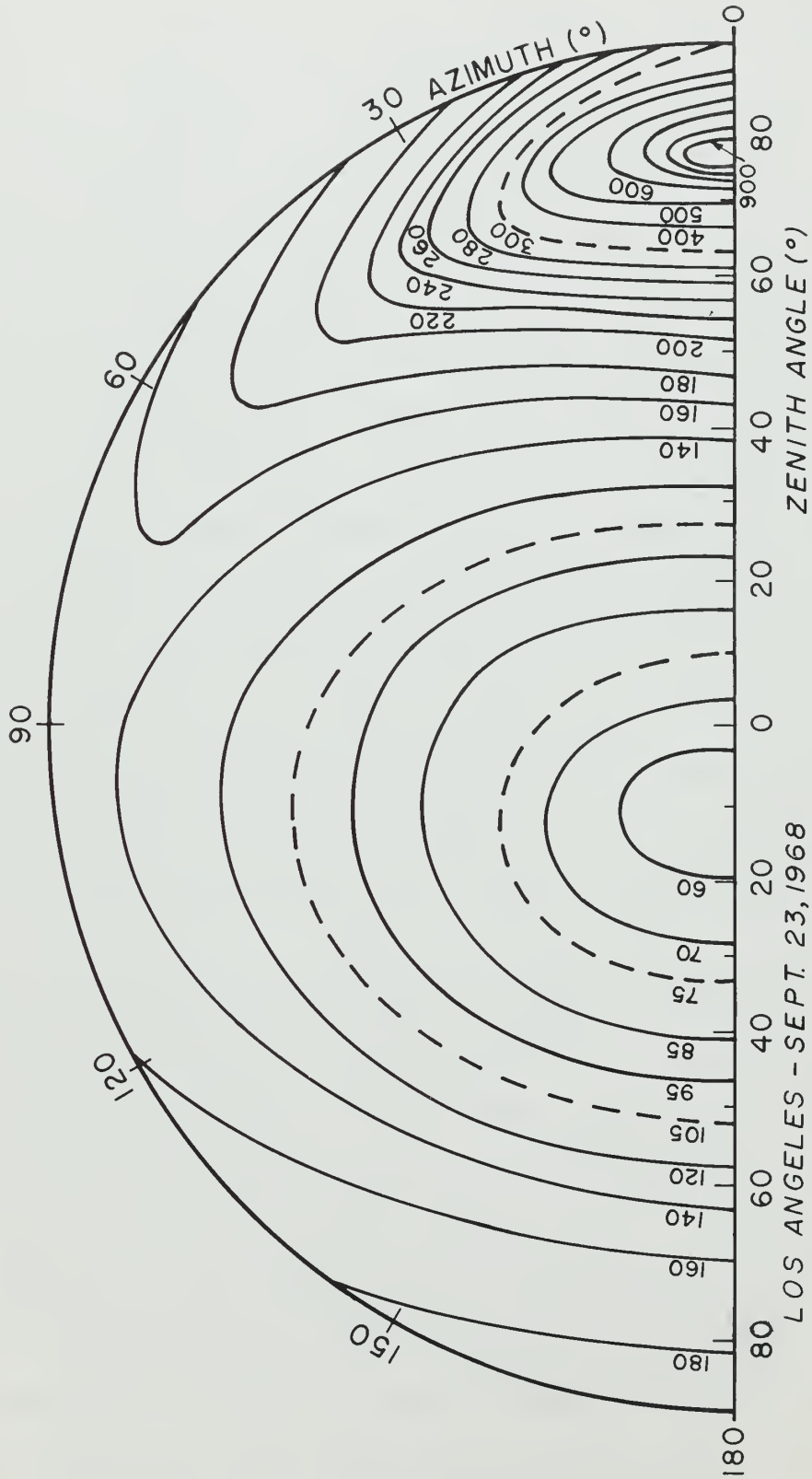


Fig. 9 Intensity distribution over the hemisphere of the sky as measured in a relatively clear atmosphere in Los Angeles (Sun elevation 14.3° , $\lambda = 3650\text{\AA}$)

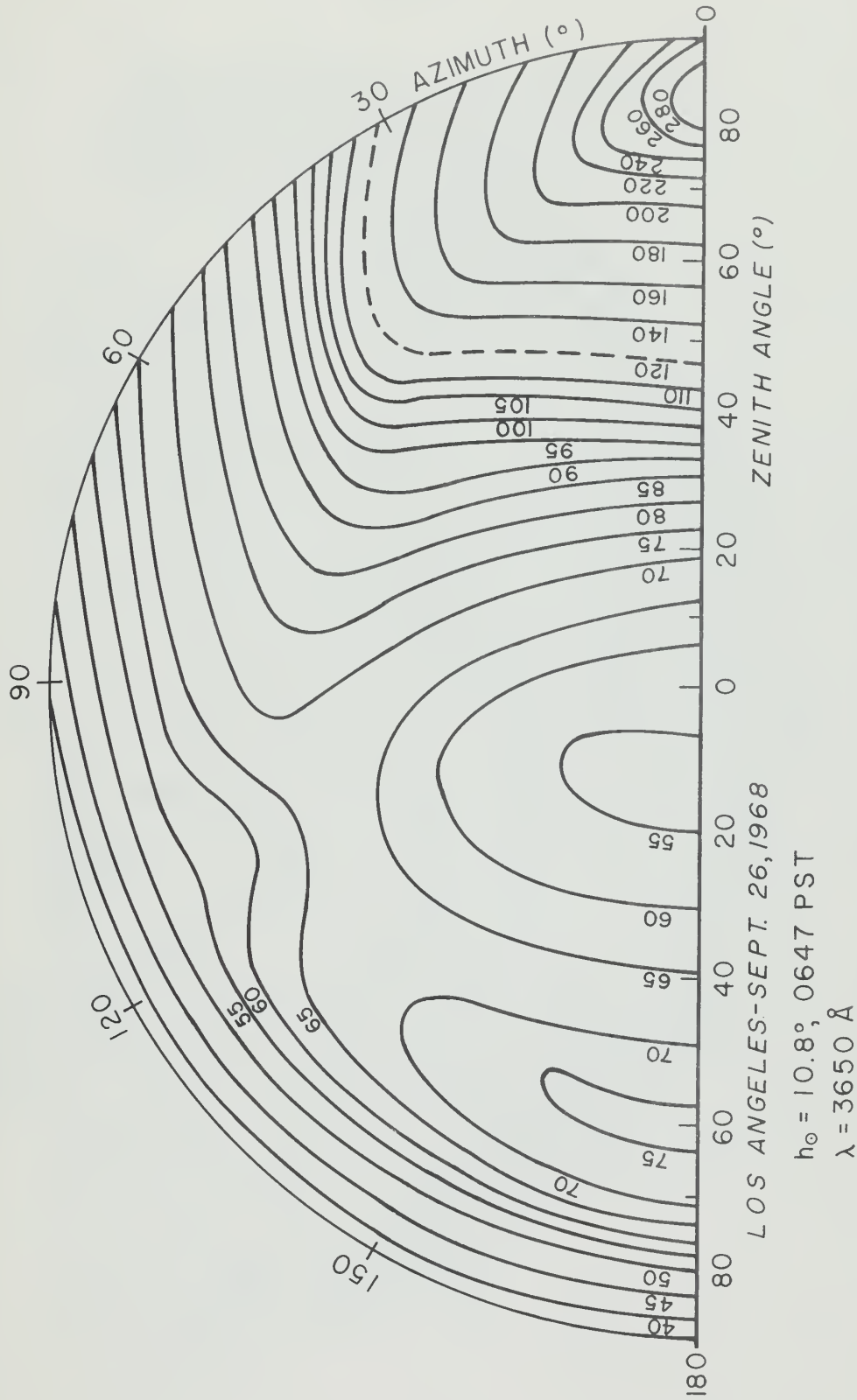
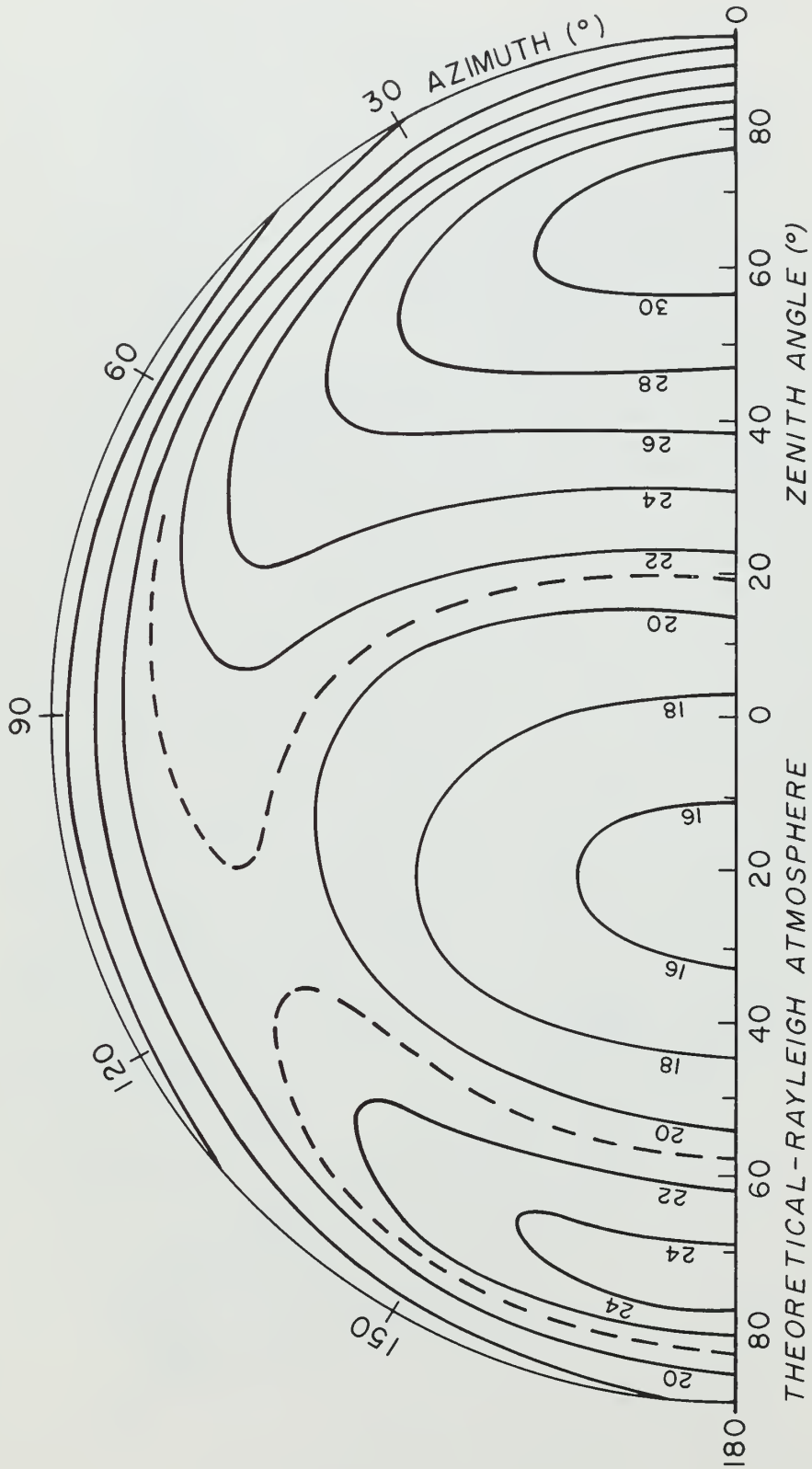


Fig. 10 Intensity distribution over the hemisphere of the sky as measured in a polluted atmosphere in Los Angeles (Sun elevation 10.8° , $\lambda = 3650\text{\AA}$)



$h_0 = 36.9^\circ$
 $\lambda = 3120 \text{ \AA}$

Fig. 11 Intensity distribution over the hemisphere of the sky as computed for a pure molecular atmosphere (Sun elevation 36.9° , $\lambda = 3120\text{\AA}$)

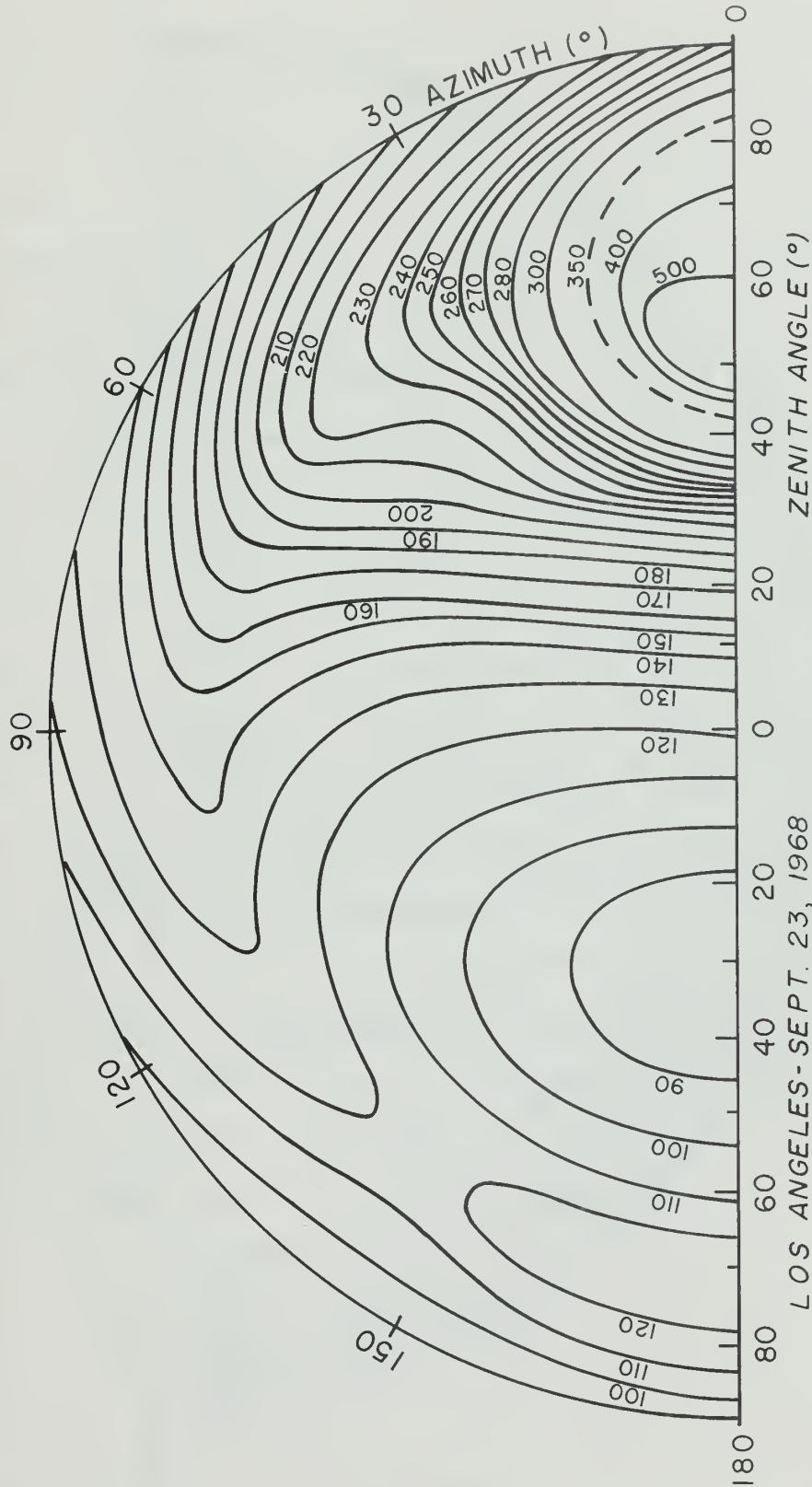
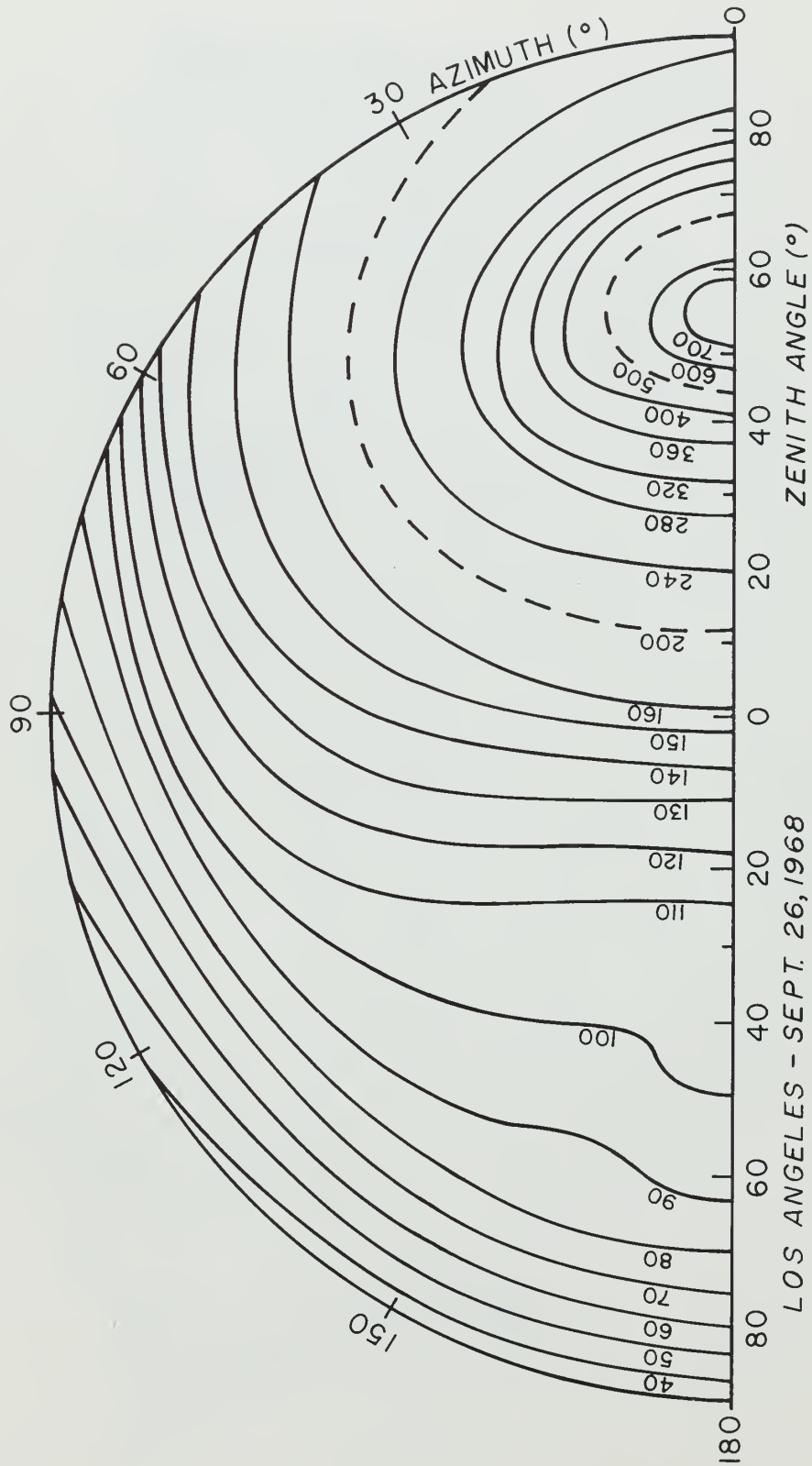


Fig. 12 Intensity distribution over the hemisphere of the sky as measured in a relatively clear atmosphere in Los Angeles (Sun elevation 36.7° , $\lambda = 3650\text{\AA}$)



$h_0 = 35.9^\circ$, 0856 PST

$\lambda = 3650 \text{ \AA}$

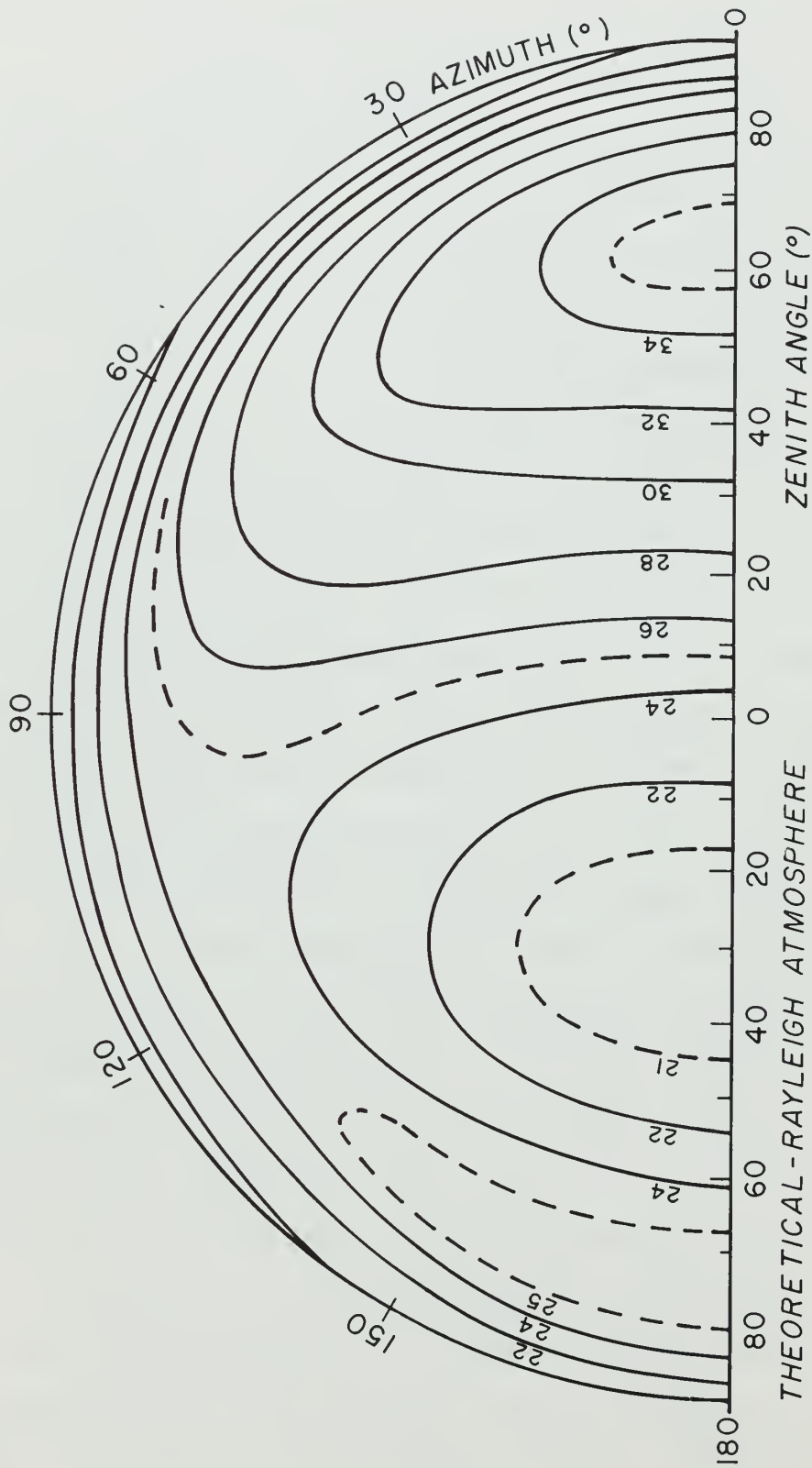
Fig. 13 Intensity distribution over the hemisphere of the sky as measured in a polluted atmosphere in Los Angeles (Sun elevation 35.9° , $\lambda = 3650\text{\AA}$)

The pattern otherwise did not change greatly except for the shift of position mentioned above.

A very considerable change of the pattern, however, occurred during the early forenoon in the case of smog, as shown by Figure 13. The minimum in the field has disappeared completely, although there is still a weakening of the intensity gradient at the $\phi = 180^\circ$ azimuth. Horizon darkening remained strong.

High sun elevation is accompanied by a further shift of the patterns for all three cases, as shown by Figures 14, 15, and 16. Aside from this shift, the Rayleigh intensity distribution in Figure 14 is similar to that in Figures 8 and 11. Figure 15 shows that the solar aureole remained strong up to the highest sun elevation reached during the measurements, and that the sky below the sun remained very bright. These characteristics are caused by relatively large numbers of Mie particles, and by solar noon they had been mixed through a relatively deep layer. The existence of the minimum reflects the Rayleigh scattering influence on the radiation field.

The most extreme smog encountered during the measurements occurred at about the time of Figure 16. In this case the sky generally appeared a bright milky white, with very little blue showing through, and the regions near the horizons had a darker brown color. As is indicated in the diagram, the brightest part of the sky was near the sun, and the intensity decreased monotonically from the sun to the horizon at all azimuths. Obviously, the scattering pattern was dominated by Mie type scatterers, and the course of the isopleths around the position of the sun indicates that the controlling parameter



$h_0 = 53.1^\circ$
 $\lambda = 3120 \text{ \AA}$

Fig. 14 Intensity distribution over the hemisphere of the sky as computed for a pure molecular atmosphere (Sun elevation 53.1° , $\lambda = 3120\text{\AA}$)

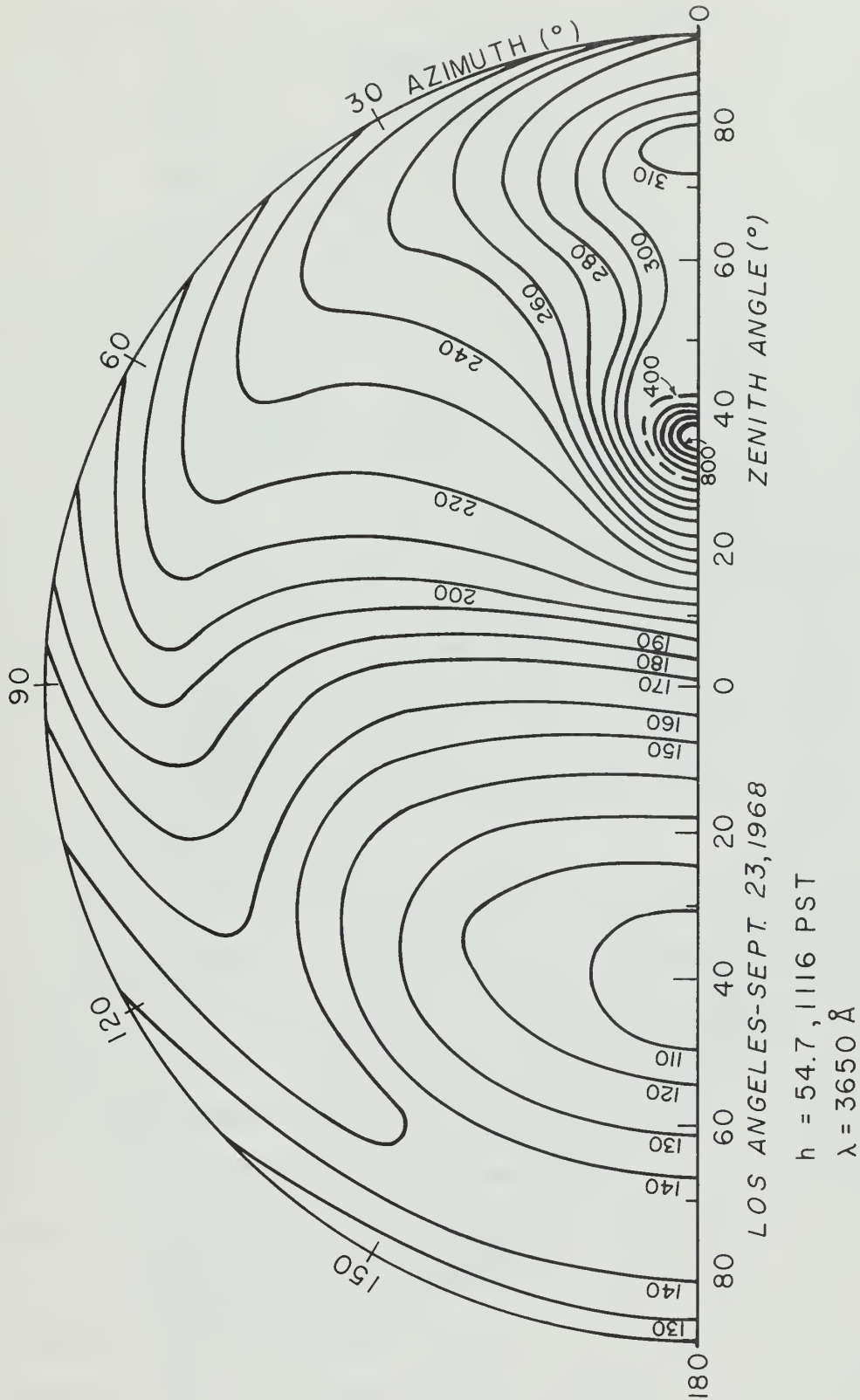


Fig. 15 Intensity distribution over the hemisphere of the sky as measured in a relatively clear atmosphere in Los Angeles (Sun elevation 54.7°, $\lambda = 3650\text{\AA}$)

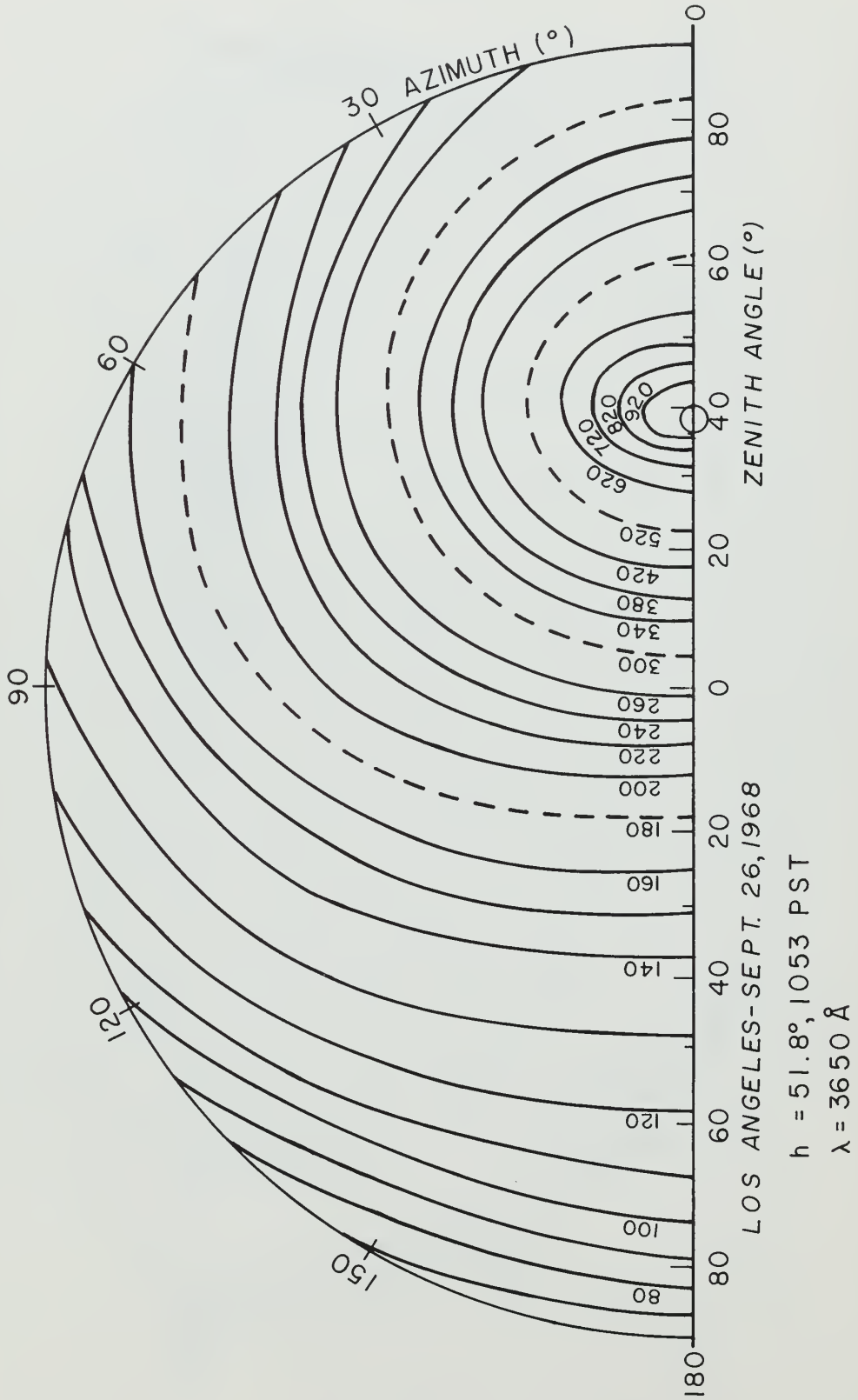


Fig. 16 Intensity distribution over the hemisphere of the sky as measured in a polluted atmosphere in Los Angeles (Sun elevation 51.8° , $\lambda = 3650\text{\AA}$)

was scattering angle taken with respect to the position of the sun. Thus, primary scattering by aerosols must have been very strong, in spite of the fact that the large concentrations of scatterers would tend to enhance the effects of multiple scattering.

In order to show the relative intensities for the two cases as explicitly as possible, curves of relative intensity in the principal plane were derived for the three selected sun elevations 10° , 30° , and 50° . The data were obtained by interpolation from the measurements. The early morning intensities applicable to a sun elevation of 10° are shown in Figure 17. The correspondence between the curves is striking, considering the quite different atmospheric conditions prevailing on the two days. This reconfirms the conclusion that the pollution particles were confined to low levels in the early morning hours. The fact that they were numerous, however, for the case of smog can be verified by the limb darkening which occurred for that case and not for the clear case.

Mid-forenoon conditions, corresponding to a sun elevation of 30° , are shown in Figure 18. A stronger increase of intensity from that shown for 10° sun elevation in Figure 17 occurred for the case of smog than for the clear case, and the effects of smog erased the broad minimum of the intensity field. Otherwise, the distributions are similar. These features were further emphasized as the smog built up, resulting in the patterns shown in Figure 19 for a sun elevation of 50° . The intensity difference is nearly a factor of two over much of the hemisphere, but the horizons remained dark in smog. The relative broadening of the aureole by smog is clearly evident. A corresponding curve for the Rayleigh atmosphere is included for comparison, the

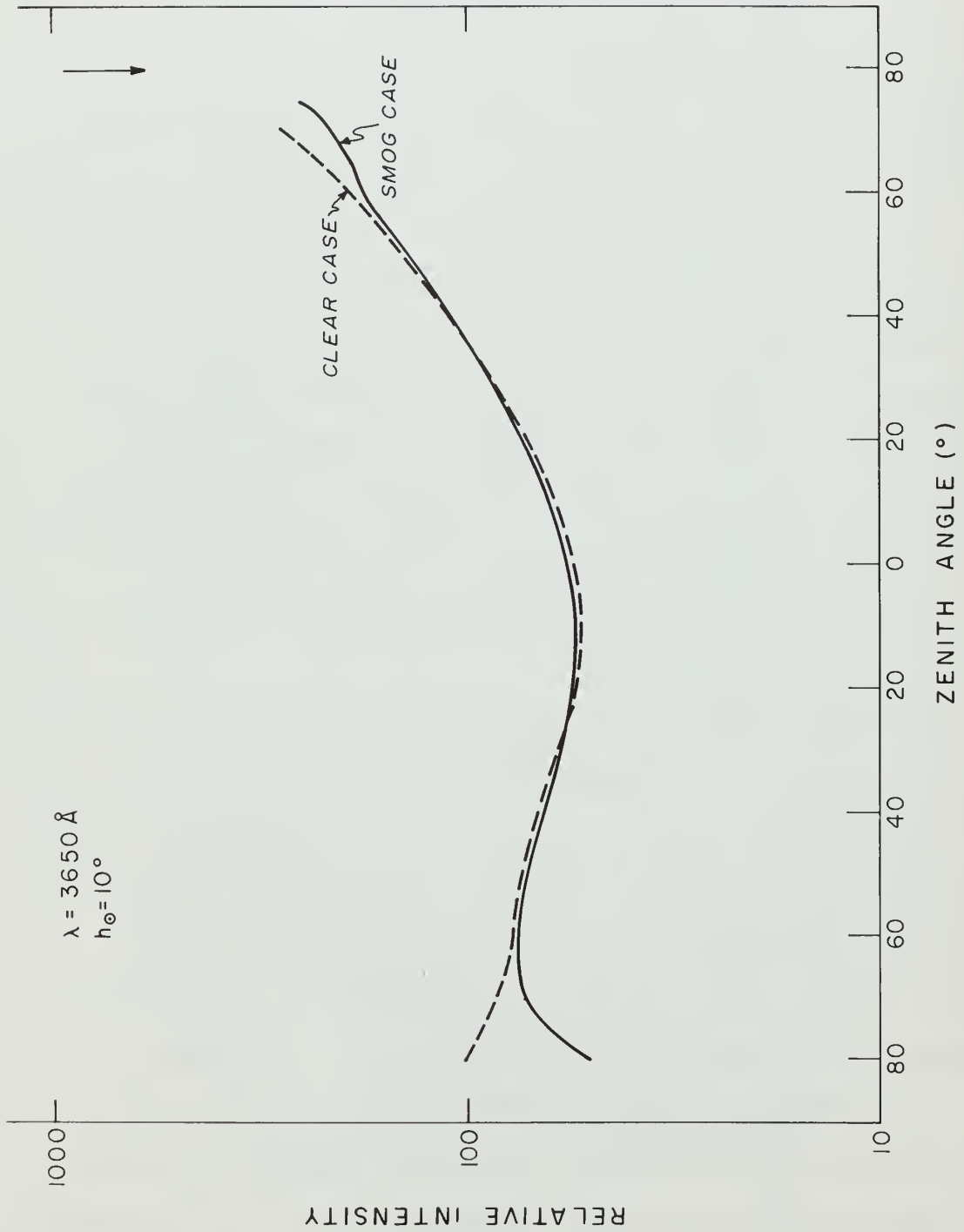


Fig. 17 Relative intensity of skylight as measured in Los Angeles for a relatively clear atmosphere and for a polluted atmosphere (Sun elevation 10° , $\lambda = 3650 \text{ \AA}$, principal plane)

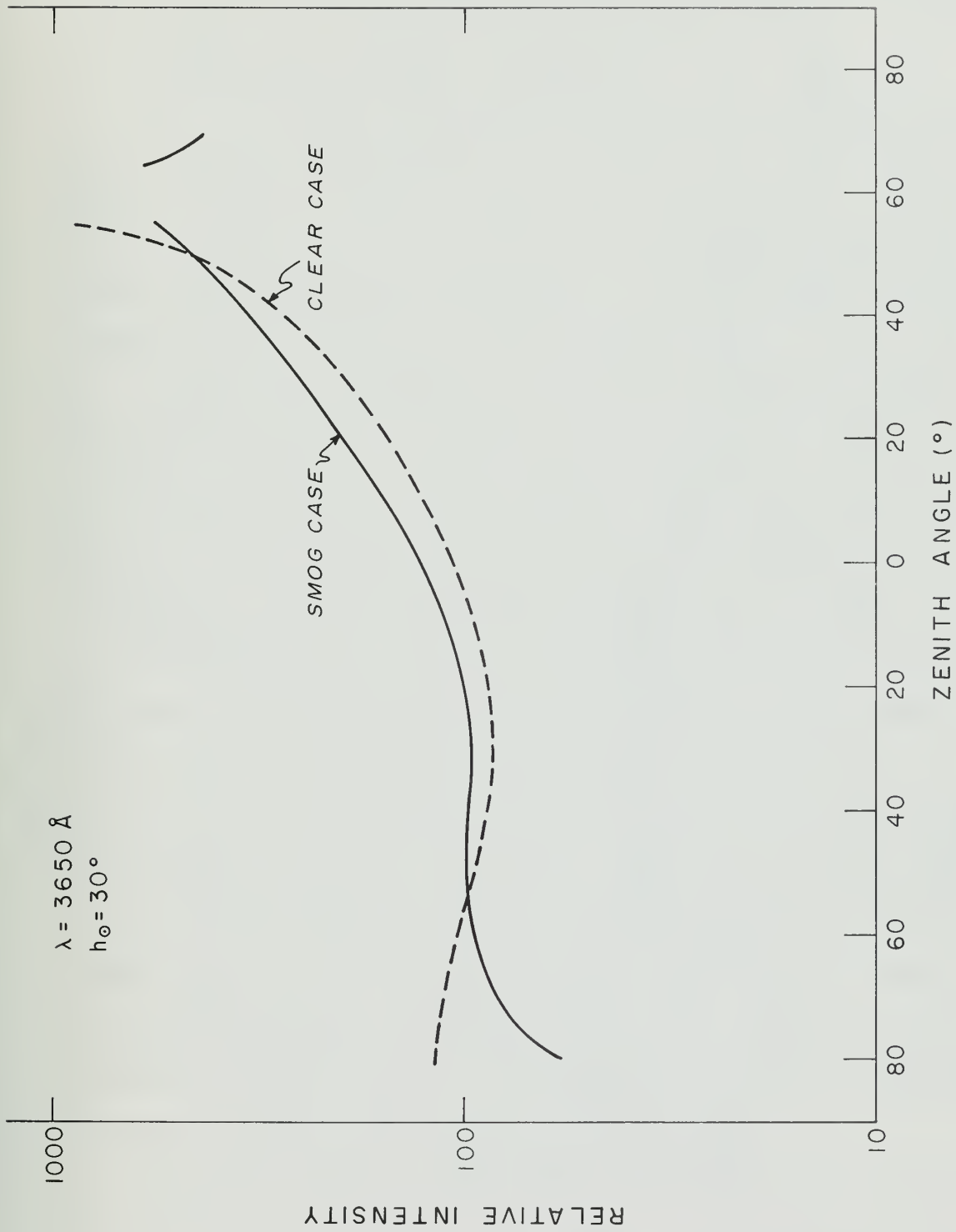


Fig. 18 Relatively intensity of skylight as measured in Los Angeles for a relatively clear atmosphere and for a polluted atmosphere (Sun elevation 30° , $\lambda = 3650\text{\AA}$, principal plane)

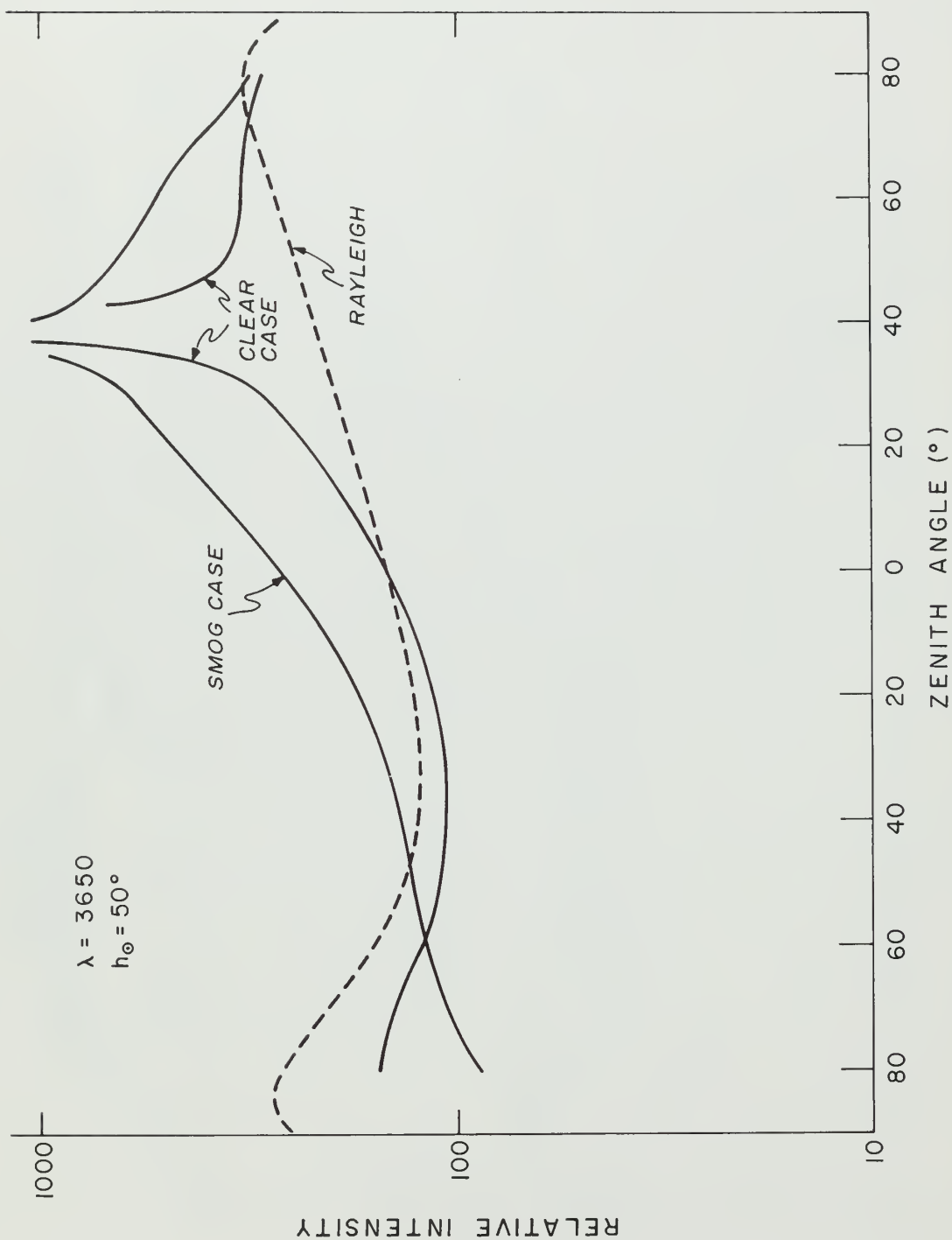


Fig. 19 Relative intensity of skylight as measured in Los Angeles for a relatively clear atmosphere, and for a polluted atmosphere, and as computed for a pure molecular atmosphere. The intensity for the Rayleigh case is normalized to that of the clear case at the zenith. (Sun elevation 50° , $\lambda = 3650\text{\AA}$, principal plane)

relative intensity for the Rayleigh case being normalized to correspond to the clear case in the direction of the zenith.

III. Measurements of Skylight Polarization

The discovery that light from the sunlit sky is partially polarized was made by the French physicist Arago in 1809. Since that time numerous authors have reported on investigations, both theoretical and observational, of the polarization of skylight. A good summary of the historical development of the subject was given by Sekera (1950).

The effect of scattering by non-Rayleigh particles was observed extensively after the volcanic eruptions of Krakatao in 1883 and Mt. Katmai in 1912. Cornu (1884) observed a decrease of the maximum polarization from 0.75 to less than 0.48 by volcanic debris from Krakatao, and Dorno (1919) found a similar effect after the eruption of Mt. Katmai. The positions of the neutral points (points of zero polarization), especially the Babinet and Arago points, were found to be particularly sensitive indicators of turbidity of the type introduced by volcanic ash.

More recent observations of polarization have been oriented toward determining characteristics of the troposphere. Stanov (1963) endeavored, with only limited success, to derive a method of determining atmospheric turbidity in various directions by polarization measurements, and Pyaskovskaya-Fessenkova (1961) investigated the relationship between atmospheric transparency and the polarization of skylight. An extensive discussion of the problem was given by Jensen (1942).

Measurements using the modern types of electronic polarimeters have been reported by Sekera (1957) and by deBary (1964) for mainly clear atmospheres. Equivalent data for the case of heavily polluted atmospheres,

however, are sparse. Perhaps the most extensive single series of such measurements is that of Holzworth and Rao (1965). They obtained data in three regions of the visible spectrum on ten different days from three locations in the Los Angeles area and analyzed the data in terms of the maximum polarization and the behavior of the neutral points.

In the present investigation, both polarization and relative intensity were obtained from the output signal from the polarizing radio-meter, so polarization data are available for the same cases as are intensities. In order to not introduce unknown effects of reflection from different locations, the emphasis here will be on polarization measurements obtained on clear and smoggy days at the same location in Los Angeles.

Curves of the degree of polarization in the principal plane* are shown for the clear atmospheric case (September 23, 1968) for $\lambda = 3200\text{\AA}$ in Figure 20 and $\lambda = 3650\text{\AA}$ in Figure 21. The general shape of the curves is essentially independent of sun elevation, and the degree of polarization at the maximum shows only a small variation with sun elevation. Both of these characteristics are typical of scattering in a Rayleigh atmosphere. However, the magnitude of the degree of polarization at the maximum is considerably less than that for the Rayleigh atmosphere, and the magnitude of the negative polarization in the region near the sun is less than that of the Rayleigh atmosphere. These characteristics, which can be seen more explicitly in later diagrams (Figures 24, 25, and 26), are in accord with those which are predicted for aerosol scattering.

* The azimuth scale was inadvertently reversed in Figures 20, 21, 22, and 23 from that in other diagrams. In these particular Figures the azimuth of the sun is in the left half of the diagram, whereas in the other Figures it is in the right half.

$\lambda = 3200 \text{ \AA}$
 SEPTEMBER 23, 1968
 LOS ANGELES CALIF.
 CLEAR CASE

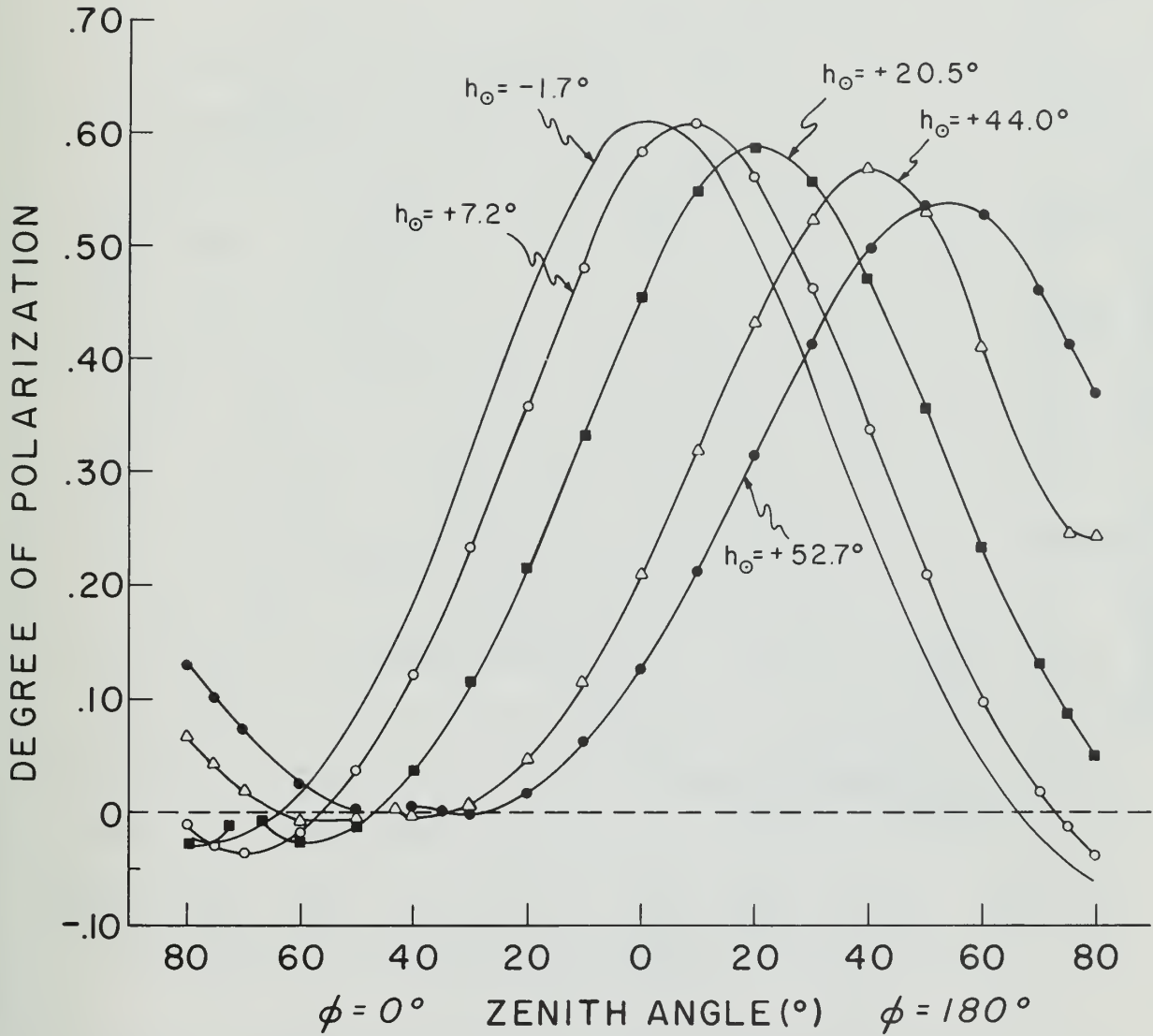


Fig. 20 Degree of polarization of skylight as measured in a relatively clear atmosphere in Los Angeles at five different sun elevations (Principal plane, $\lambda = 3200\text{\AA}$)

$\lambda = 3650 \text{ \AA}$
 SEPTEMBER 23, 1968
 LOS ANGELES CALIF
 CLEAR CASE

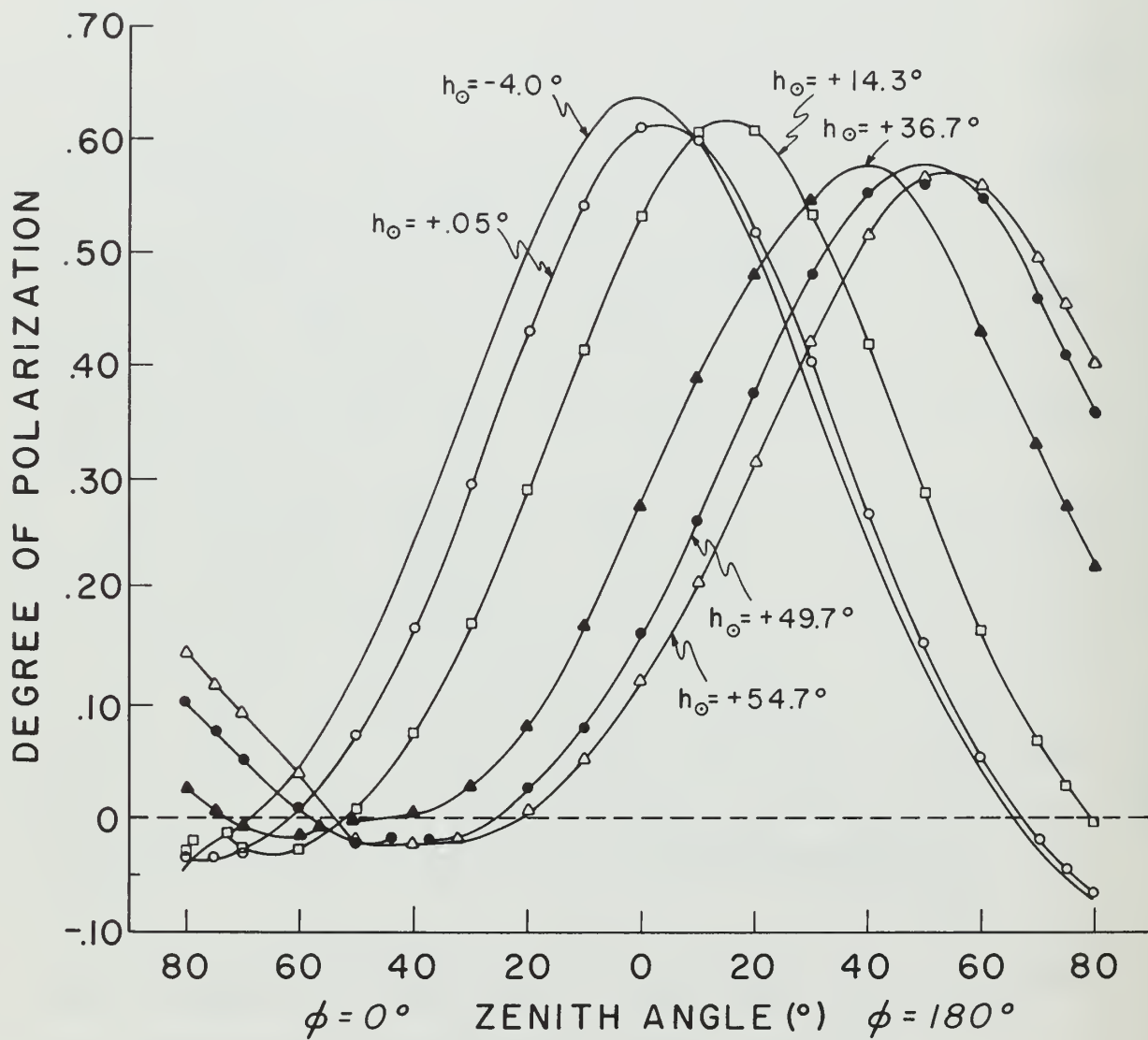


Fig. 21 Degree of polarization of skylight as measured in a relatively clear atmosphere in Los Angeles at six different sun elevations (Principal plane, $\lambda = 3650\text{\AA}$)

The equivalent diagrams for the case of smog (September 26, 1968) in Los Angeles are shown in Figures 22 and 23. The general shape of the curves is not greatly altered by smog, but the magnitudes of both the degree of polarization at the maximum and of the negative polarization near the sun are greatly decreased by the effects of aerosol scattering. The variation with solar elevation is brought about by a combination of the increase of density of the aerosol particles and the increase of thickness of the smog layer with increasing sun elevation. While it is not particularly obvious from the diagram, there was a progressive shift of the positions of the maximum with respect to the sun from approximately 93° from the sun before sunup to about 85° near solar noon. It will be remembered that the maximum is at exactly 90° from the sun for primary Rayleigh scattering. Fraser (1955) found that a shift similar to that observed here can be expected to occur at short wavelengths in a Rayleigh atmosphere, but it does not seem to be explained by the theory for aerosol scattering. For instance, van de Hulst (1957) indicates that scattering by Mie particles causes the maximum to occur at scattering angles greater than 90° , and Deirmendjian (1962) shows the maximum polarization for scattering of blue light by haze to occur at a scattering angle of 100° . Similar results have been obtained by Coulson (unpublished computations) for an atmosphere which is characterized by Junge's (1963) continental model of atmospheric turbidity. However, a shift to angles less than 90° was observed by Fraser (1955) to occur at short wavelengths in natural atmospheric conditions at several locations, in corroboration of the measurements obtained in this study.

$\lambda = 3200 \text{ \AA}$
 SEPTEMBER 26, 1968
 LOS ANGELES CALIF.
 SMOG CASE

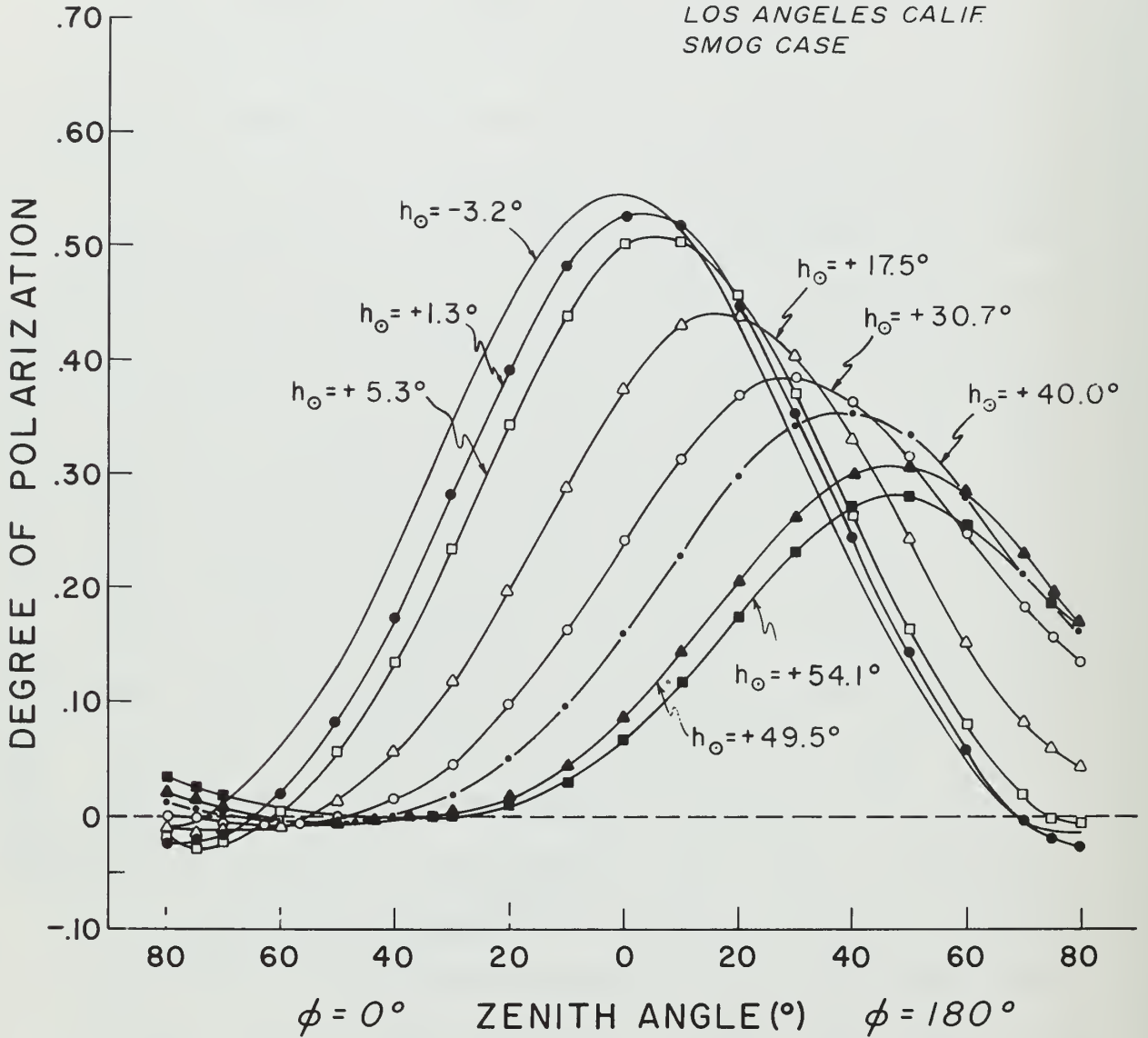


Fig. 22 Degree of polarization of skylight as measured in a polluted atmosphere in Los Angeles at seven different sun elevations (Principal plane, $\lambda = 3200\text{\AA}$)

$\lambda = 3650 \text{ \AA}$
 SEPTEMBER 26, 1968
 LOS ANGELES CALIF.
 SMOG CASE

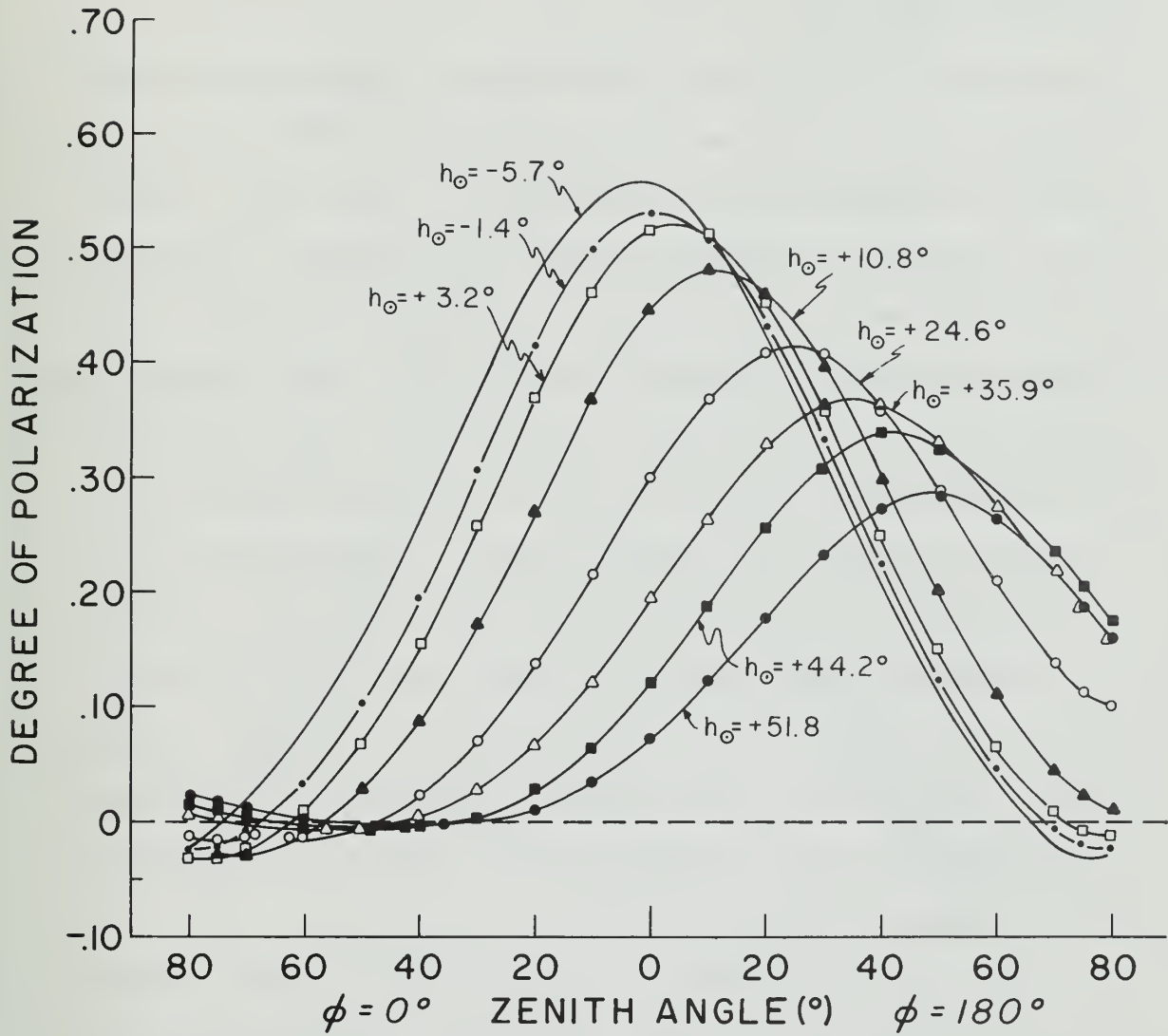


Fig. 23 Degree of polarization of skylight as measured in a polluted atmosphere in Los Angeles at eight different sun elevations (Principal plane, $\lambda = 3650\text{\AA}$)

The curves of Figures 24, 25, and 26 show somewhat more explicitly the effect of smog on the polarization field of skylight at three different sun elevations, 10° , 30° , and 50° . Interpolation among the data was necessary in some cases to obtain curves at these particular positions of the sun. These results are only for $\lambda = 3650\text{\AA}$, but they are generally representative of measurements at $\lambda = 3200\text{\AA}$ also. Curves for the Rayleigh atmosphere are included for comparison. The most important modification of the polarization field by smog are (a) a pronounced decrease of polarization throughout most of the principal plane, including both positive and negative areas of the field, (b) a small but consistent shift of the positions of the neutral points, in the sense that the Babinet and Brewster points are shifted toward the sun and the Arago point is shifted toward the anti-sun, and (c) a shift of the position of the polarization maximum from about 90° from the sun at a sun elevation of 10° to less than 90° at the higher sun elevations. The first two of these effects are consistent with what would be expected from the Mie scattering theory, while the last one is not.

IV. Positions of the Neutral Points

As mentioned previously, it has been known for nearly a century that the positions of the neutral points of skylight polarization, particularly the Babinet and Arago points, are sensitive indicators of atmospheric turbidity. The existence of volcanic ash in the atmosphere produces large changes of neutral point behavior (Cornu - 1884, Dorno - 1919), the particular type of behavior observed after major volcanic eruptions apparently being produced only by volcanic ash. However, measurements at other times show a sensitivity of neutral point positions

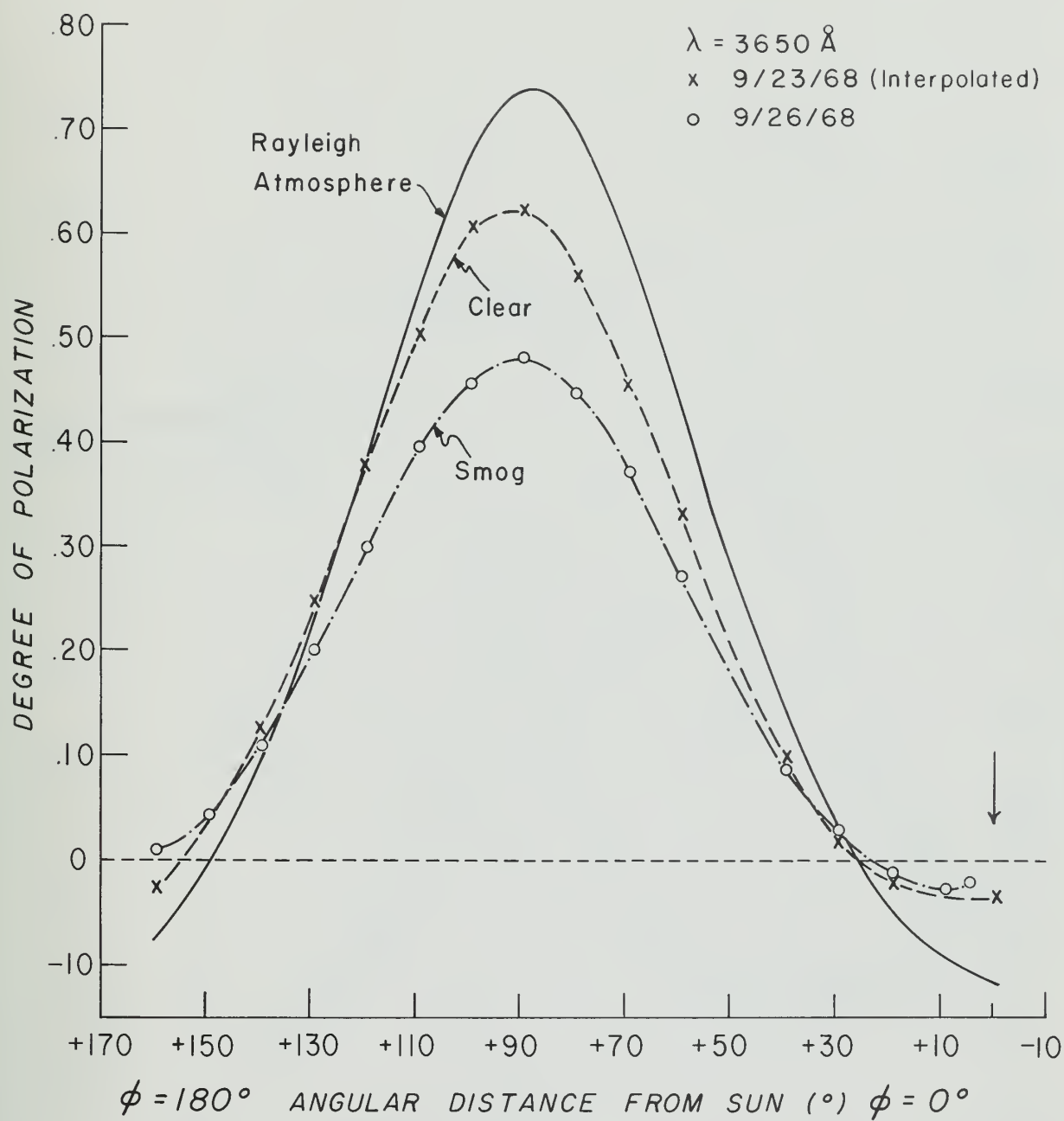


Fig. 24 Degree of polarization of skylight as measured in Los Angeles for a relatively clear atmosphere and for a polluted atmosphere, and as computed for a pure molecular atmosphere. (Sun elevation 10° , $\lambda = 3650\text{\AA}$, principal plane)

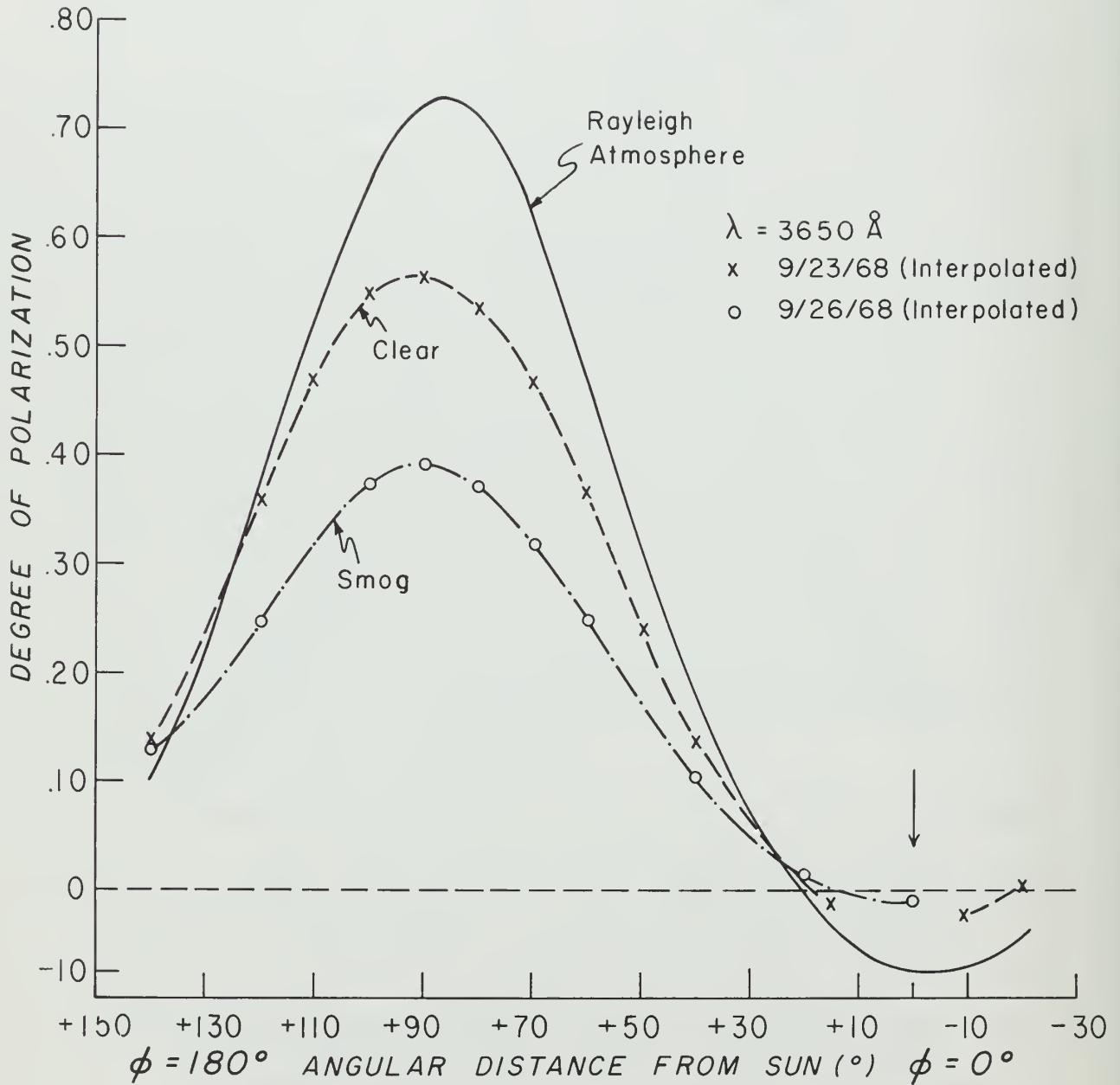


Fig. 25 Degree of polarization of skylight as measured in Los Angeles for a relatively clear atmosphere and for a polluted atmosphere, and as computed for a pure molecular atmosphere (Sun elevation 30° , $\lambda = 3650\text{\AA}$, principal plane)

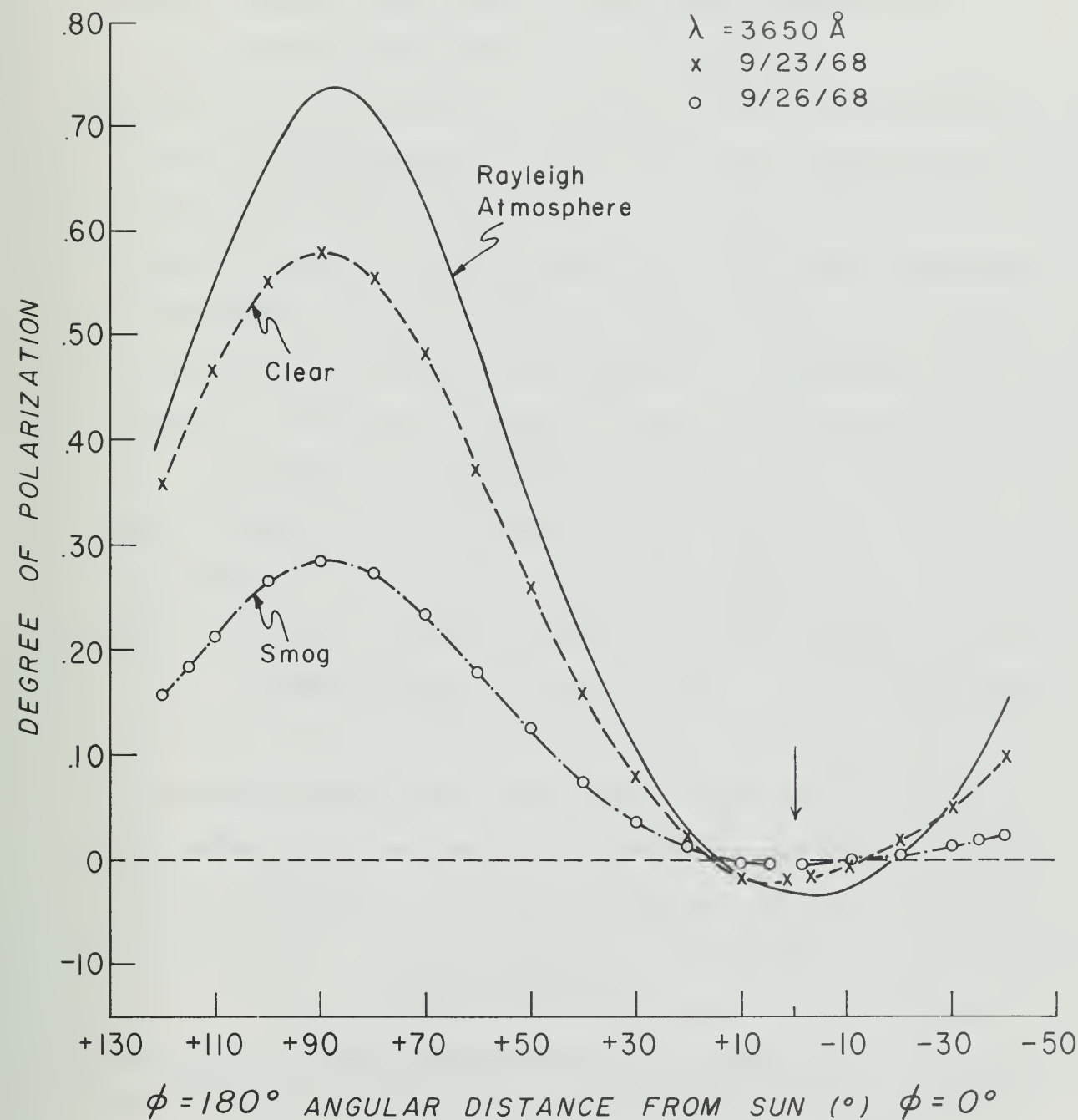


Fig. 26 Degree of polarization of skylight as measured in Los Angeles for a relatively clear atmosphere and for a polluted atmosphere, and as computed for a pure molecular atmosphere (Sun elevation 50° , $\lambda = 3650\text{\AA}$, principal plane)

to other types of aerosol scattering as well, although the reaction is not as specific for other types of particles as for volcanic ash.

A long series of observations of neutral point positions was made by Sekera and Collaborators (1955). Emphasis there, however, was on selected cases of a clear atmosphere, and not on the polluted atmosphere of main interest in the present study. Neuberger (1950) discussed the effect of turbidity on the position of the Arago point, but up until the recent development of electronic polarimeters all of the measurements were by visual instrumentation, and thus were confined to the visible wavelengths. As mentioned above, Holzworth and Rao (1965) used an electronic polarimeter for obtaining measurements of neutral points at three wavelengths in visible region of the spectrum.

It has become clear from the available data in the blue and ultraviolet regions that the usual effect of large particle scattering is to shift the positions of the Babinet and Brewster points in the direction toward the sun. The Arago point is sometimes observed to be shifted farther from the anti-sun and sometimes toward the anti-sun (Holzworth and Rao - 1965). These same types of shifts are seen to occur in the present measurements of the position of the Babinet point in Figures 27 and 28. Equivalent data for the Brewster point are too sparse in these measurements for adequate analysis, but they will be presented in a later report. The behavior of the Arago point is more regular than that observed by Holzworth and Rao (1965), as can be seen by Figures 29 and 30. In these measurements, the Arago point is definitely shifted toward the anti-sun by aerosol scattering effects. Only at negative sun elevations is there some uncertainty in the observed behavior.

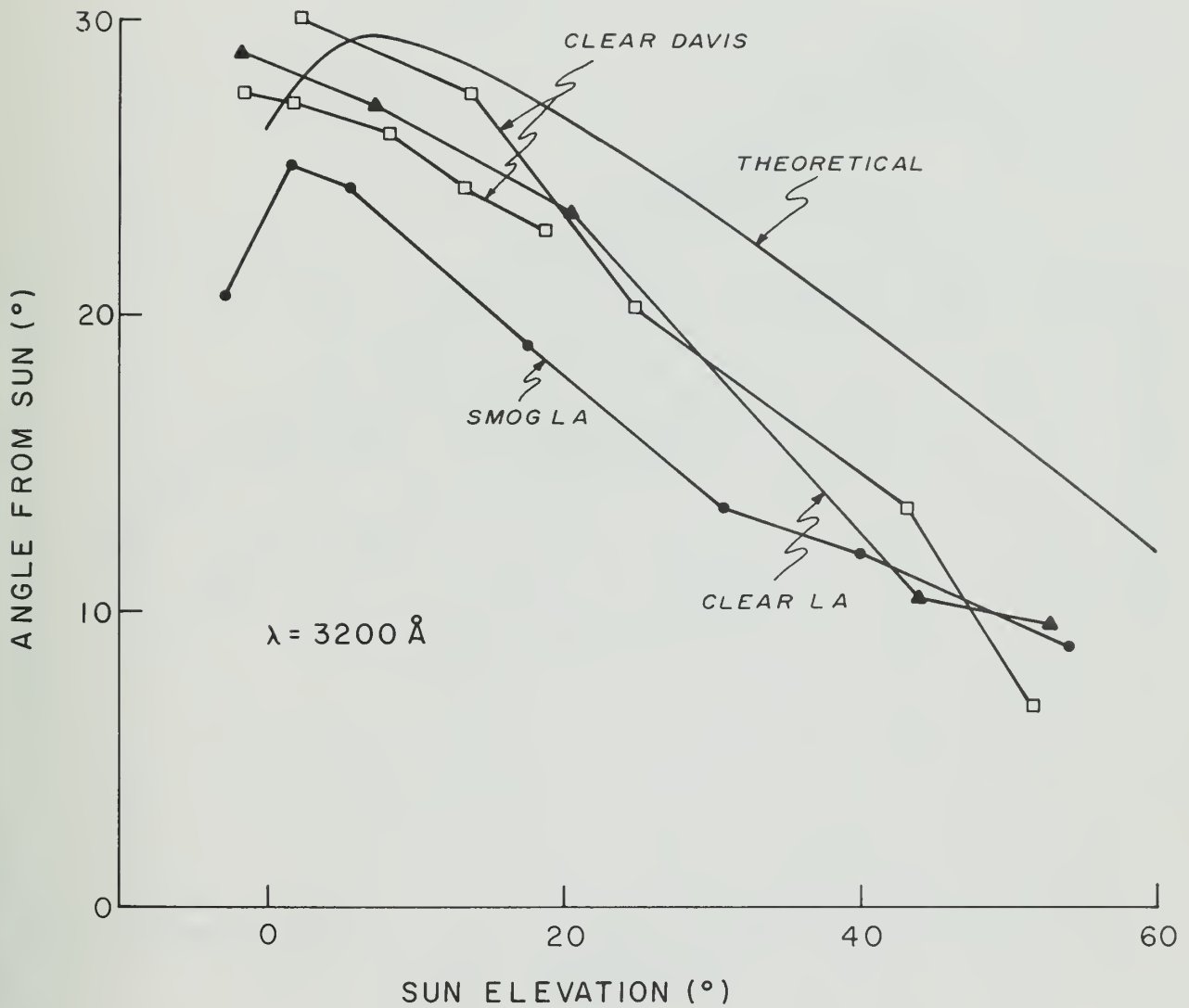


Fig. 27 Angular distance of the Babinet point from the sun as measured in clear atmospheres in Los Angeles and Davis and in a polluted atmosphere in Los Angeles, and as computed for a pure molecular atmosphere ($\lambda = 3200\text{\AA}$)

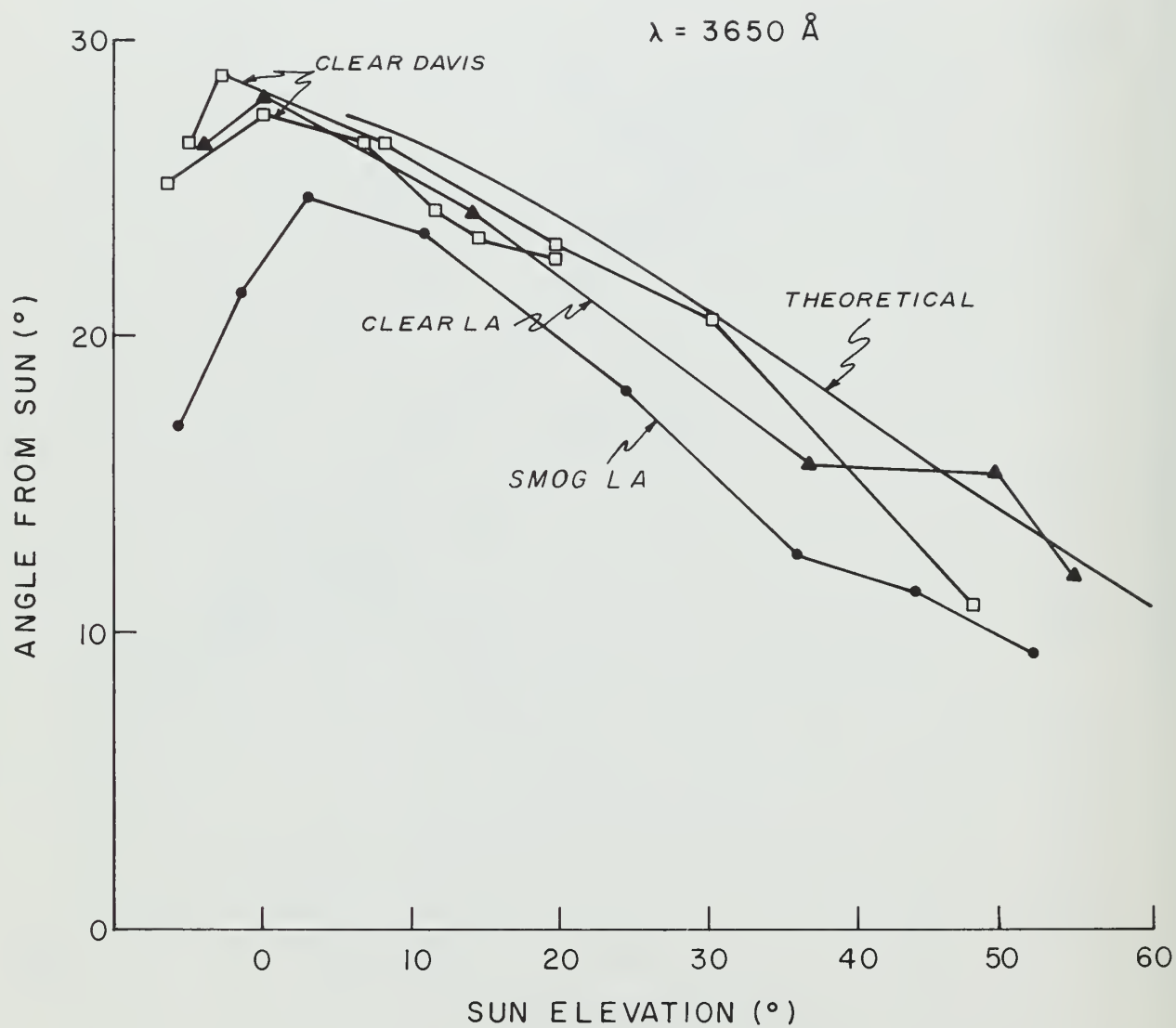


Fig. 28 Angular distance of the Babinet point from the sun as measured in clear atmospheres in Los Angeles and Davis and in a polluted atmosphere in Los Angeles, and as computed for a pure molecular atmosphere ($\lambda = 3650\text{\AA}$)

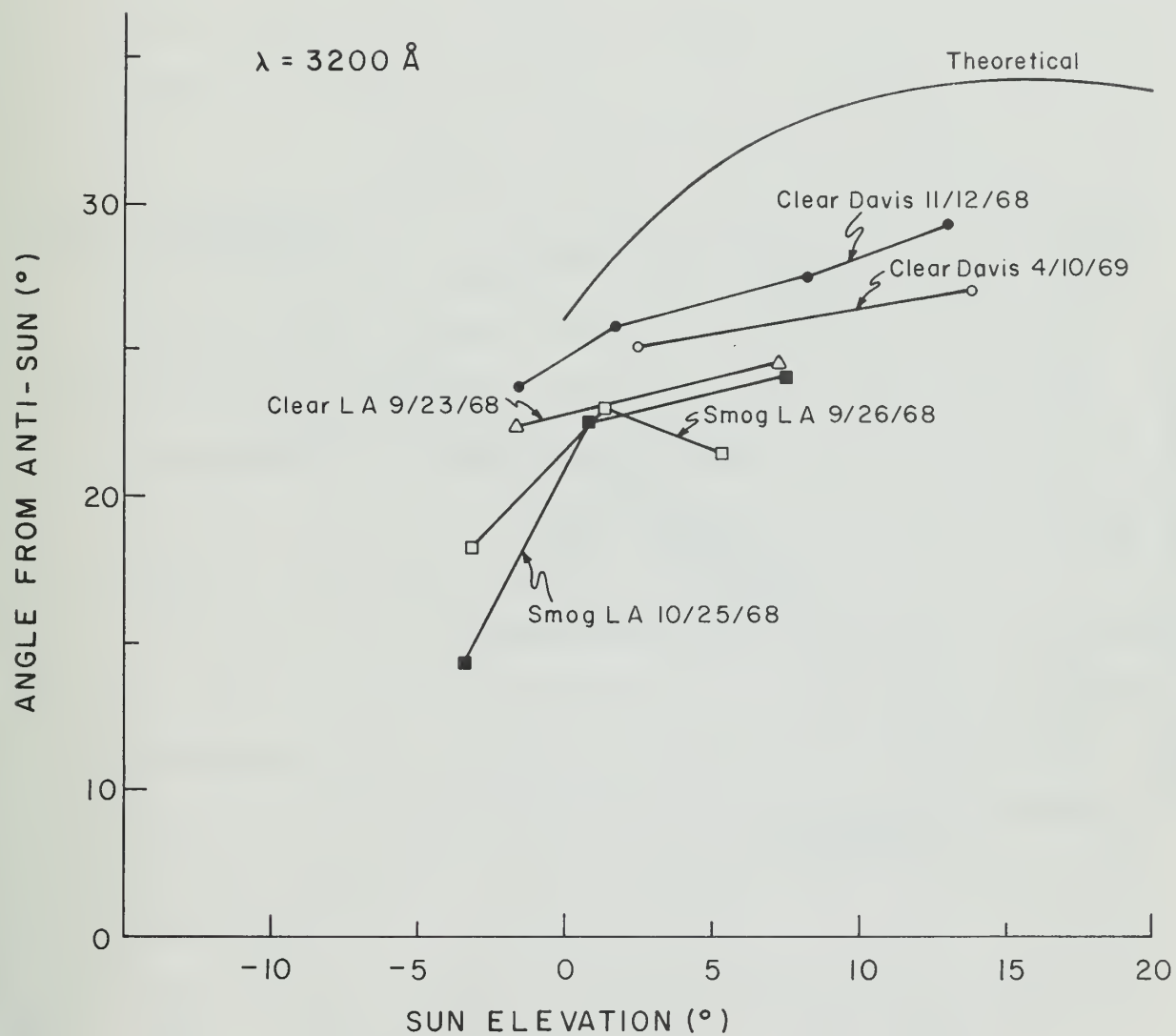


Fig. 29 Angular distance of the Arago point from the anti-sun as measured in clear atmospheres in Los Angeles and Davis and in a polluted atmosphere in Los Angeles, and as computed for a pure molecular atmosphere ($\lambda = 3200\text{\AA}$)

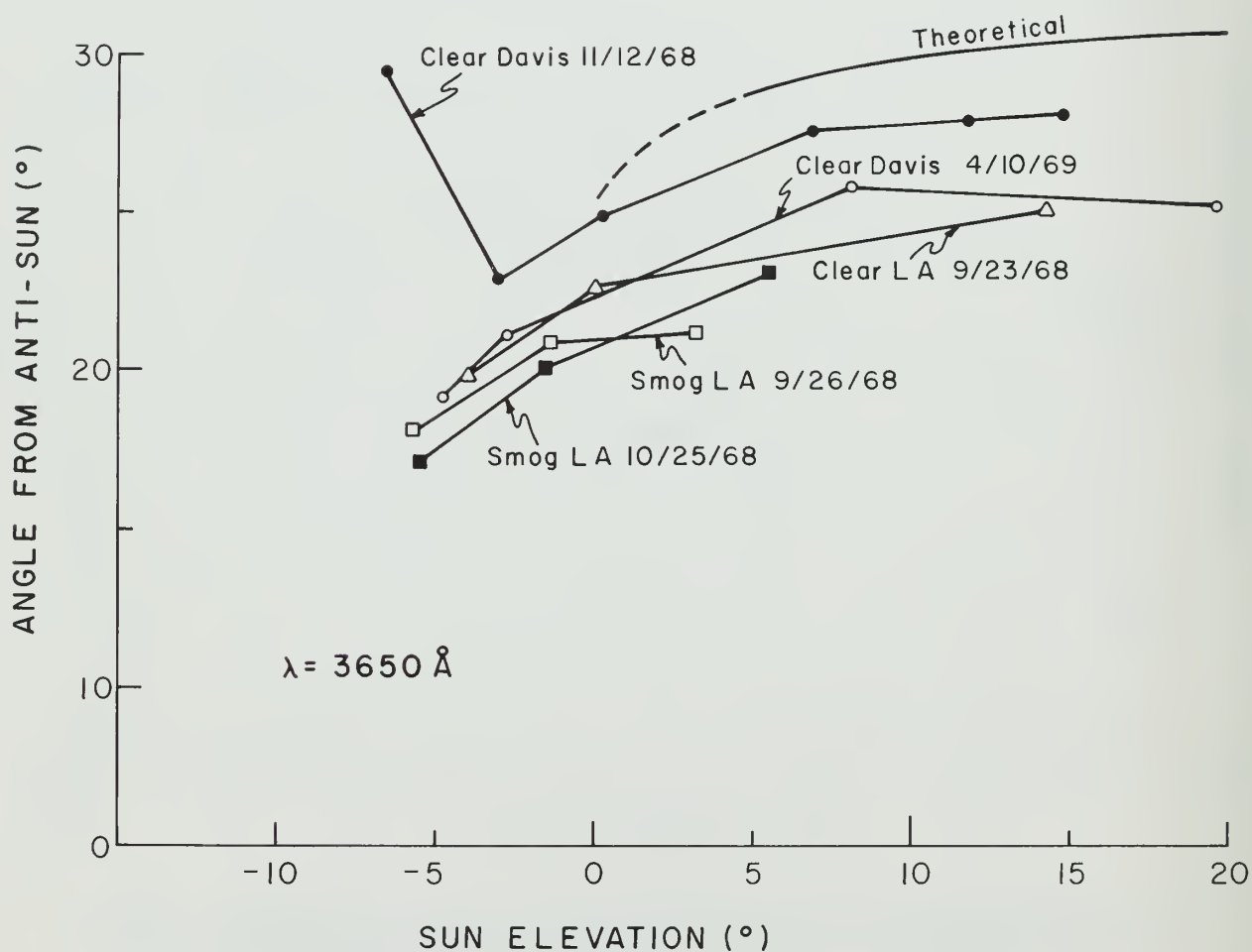


Fig. 30 Angular distance of the Arago point from the anti-sun as measured in clear atmospheres in Los Angeles and Davis and in a polluted atmosphere in Los Angeles, and as computed for a pure molecular atmosphere ($\lambda = 3650 \text{ \AA}$)

The computed behavior of the Babinet point for a clear atmosphere is shown by the theoretical curves of Figures 27 and 28. The neutral point approaches the sun with increasing sun elevation, and for the sun at the zenith both the Babinet and Brewster points would coincide with the sun. It is obvious from symmetry relations that this must be the case, and computations for a Rayleigh atmosphere verify the fact (Coulson, 1954). The theory also shows the angle of the neutral point from the sun to reach a maximum* at a few degrees from sun elevation. Both of these characteristics are reproduced reasonably well by the measurements, although the maximum was not reached for the clear atmospheric cases at $\lambda = 3200\text{\AA}$. It shows up well for all measurements at $3650\text{\AA}$, however, and for the polluted case at $3200\text{\AA}$. The larger angles for the observed Babinet point than the theoretical for low sun elevations is in accord with the theoretical behavior reported by Kano (1968) for an atmosphere containing a low level turbid layer.

The general behavior of the Babinet point can be seen to correspond well to what has been observed previously and to what would be expected from scattering theory. Positive polarization is introduced into the radiation field by aerosol scattering, the more aerosols present the more positive polarization which is produced at these scattering angles. It is not completely obvious that such should be the case, particularly for heavy pollution. Multiple scattering effects become extremely important in that case, and multiple scattering tends to depolarize the radiation field. It is realized, of course, that the field near the neutral points has very little polarization, so the addition of even

* The computations necessary to extend the theoretical curve for $\lambda = 3650\text{\AA}$ to 0° sun elevation have not been completed. However, curves for wavelengths both above and below $3650\text{\AA}$ show the maximum, so it is logical that the maximum would exist at $3650\text{\AA}$ also.

a very small amount of positively polarized light can cause a significant shift of the position of the neutral point. This fact accounts for the observed sensitivity of neutral point positions to atmospheric aerosols.

Neuberger (1950) concluded that the effects of turbidity on the position of the Arago point is most consistently evident in the magnitude of the distance of the Arago point from the sun at the maximum, which was observed by Neuberger to occur at a sun elevation of between 5.5° and 20.5° . This criterion does not apply well to the curves of Figures 29 and 30, however, as no well defined maximum exists in the range of sun elevations for which measurements were made. In fact, by inspection of the data, it is doubtful that the maximum discussed by Neuberger is likely to be as well defined in these data as he found in his analysis. The reason for this is not clear, although the fact that his measurements were obtained for visible light whereas these are in the ultraviolet may be at least partially responsible for the discrepancy.

In most other respects the curves of Figures 29 and 30 correspond roughly to the data obtained by other observers (Sekera and Collaborators - 1955). The Arago point is farther from the anti-sun for the clear atmosphere than for the polluted atmosphere. The amplitude of the variation appears larger in the polluted case at 3200A, which is in accord with Neuberger's measurements, but this feature does not appear to be present at 3650A. The strong increase of angle reported by Neuberger to occur at negative values of sun elevation beginning at about -1° was observed in only one case ($\lambda = 3650\text{A}$, 11/12/68),

although several measurements were made at negative sun elevations of up to -5° . The reason for these differences in behavior is not known at the present time. The scattering theory which has been generally applied is for a plane parallel model of the atmosphere, in which case negative sun elevations are not considered. Once the theory is applied to the case of the spherical atmosphere it may be possible to explain the observed neutral point behavior at negative sun elevations.

V. Total Ultraviolet Flux

The flux of ultraviolet radiation through a horizontal surface has been measured by several different devices in clear and polluted atmospheres in Los Angeles during a cooperative project reported by Nader (1967). In one project, three Eppley ultraviolet pyranometers were fitted with filters to measure ultraviolet flux in one broad band and two narrow bands in the ultraviolet. Another instrument yielded flux measurements in nine narrow bands and one broad band, also in the ultraviolet region, while an integration of the effects of ultraviolet flux were performed with photosensitive papers and photosensitive plastic. Finally, one experiment using two different types of chemical actinometers was used to determine the ultraviolet flux through a specified volume of the atmosphere. In a more recent study, Nader and White (1968) developed a device for measuring the flux of ultraviolet radiation through a volume of the atmosphere in the spectral interval $3000\text{\AA} \leq \lambda \leq 3800\text{\AA}$, and used it to compare measurements of the flux through a horizontal surface with that to which a volume of the atmosphere is subjected.

As mentioned in the Introduction of this report, one of the objectives for the 1969 field trip to Los Angeles was to obtain corroborative

data on the effects of smog on the ultraviolet fluxes through horizontal and spherical surfaces. Although not all of those data are reduced at the present time, preliminary results indicate that some instrumentation problems which were not detected during the period of measurements make the data of questionable accuracy. As a consequence, we are reserving discussion of the ultraviolet flux measurements for a future report.

VI. Conclusions

On the basis of the measurements made so far, we can substantiate the following conclusions regarding ultraviolet solar radiation in clear and polluted atmospheres.

- A. Skylight intensity over most of the sky hemisphere is increased very significantly by aerosol scattering in a polluted atmosphere. The increase, being a factor of two or more, appears to be most pronounced at high sun elevations.
- B. The region of the sky near the horizons is darker, by a factor of up to two, in a polluted than in a clear atmosphere.
- C. The distribution of intensity over the sky is modified strongly by air pollution, the dominant parameter in that case being angular distance from the sun. For moderate and high sun elevations the intensity decreases monotonically with increasing angle from the sun at all azimuths.
- D. The solar aureole (increase of intensity near the sun) is not as sharp in a polluted atmosphere as in a nominally clear atmosphere.
- E. A solar aureole would not exist in a pure Rayleigh atmosphere, so there is a significant aerosol component in the clearest natural atmosphere in which measurements were made.

F. The degree of plane polarization over most of the hemisphere of the sky is decreased strongly by atmospheric pollution, the decrease at the maximum, for instance, being a factor of more than two for moderate to heavy pollution. The general pattern of polarization field is not changed a great deal by pollution, however.

G. The degree of polarization in the clearest atmosphere for which measurements were made being significantly less than that for a pure Rayleigh atmosphere indicates a significant aerosol component in the clear natural atmosphere.

H. The magnitude of the negative polarization field, although small, is more sensitive to aerosol scattering than is that of the positive part of the polarization field.

I. The position of the Babinet point is a relatively sensitive indicator of pollution, its position being shifted toward the sun by 2° to 5° or more in a polluted atmosphere from its position in a clear atmosphere.

J. The Arago point is shifted toward the anti-sun by the effects of pollution. However, the measurements so far indicate the shift of the Arago point to be somewhat smaller than that of the Babinet point under the same conditions.

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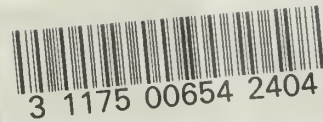
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